## IV.C Chesapeake Bay

The Chesapeake Bay, largest of the 130 estuaries in the United States, was the first in the nation to be targeted for restoration as an integrated watershed, airshed, and ecosystem. The 166,000 $km^2$ drainage basin (or watershed), shown in Figure IV-8, covers parts of six states (Delaware, Maryland, New York, Pennsylvania, Virginia, and West Virginia) and the District of Columbia, and includes more than 150 rivers and streams. The major tributary basins within the Chesapeake Bay watershed are shown in Figure IV-9, which is found later in this section.

Stretching from Havre de Grace, Maryland, to Norfolk, Virginia, the Chesapeake Bay is 314 km long, and ranges from 5 to 56 km wide. The Bay has over 90,000 km of shoreline (more than the entire West Coast of the continental United States) and a surface area of approximately 30,800 $km^2$. Generally shaped like a shallow tray, the Bay's average depth, including all tidal tributaries, is only 6 meters, with a few deep troughs running along much of its length that average 18 to 21 meters, and reaching 53 meters at the deepest point. To visualize the relatively large watershed in contrast to the small Bay volume, imagine that the Bay's watershed is reduced to the size of this page; the relative size of the Chesapeake Bay would be a section 7.2 inches by 0.9 inches in the lower right hand corner, and the average depth of the Bay would be represented by one sixtieth the thickness of the paper (see also Figure IV-8).

**Economic Highlights of Chesapeake Bay**

- In 1992, the dockside value of commercial shellfish and finfish harvests from Chesapeake Bay was close to $80 million.
- In 1993, more than 175,000 pleasure craft (e.g., sail boats) were registered in the Bay.
- Close to 1 million anglers in Maryland and Virginia made an estimated 600,000 fishing trips in 1991. Recreational fishing in these states is estimated at more than $1 billion annually.
- The Chesapeake is a key commercial waterway, and home to two of the nation's five major North Atlantic ports (Port of Baltimore, MD, and Hampton Roads Complex, VA). More than 90 million tons of cargo were shipped via the Bay in 1992.

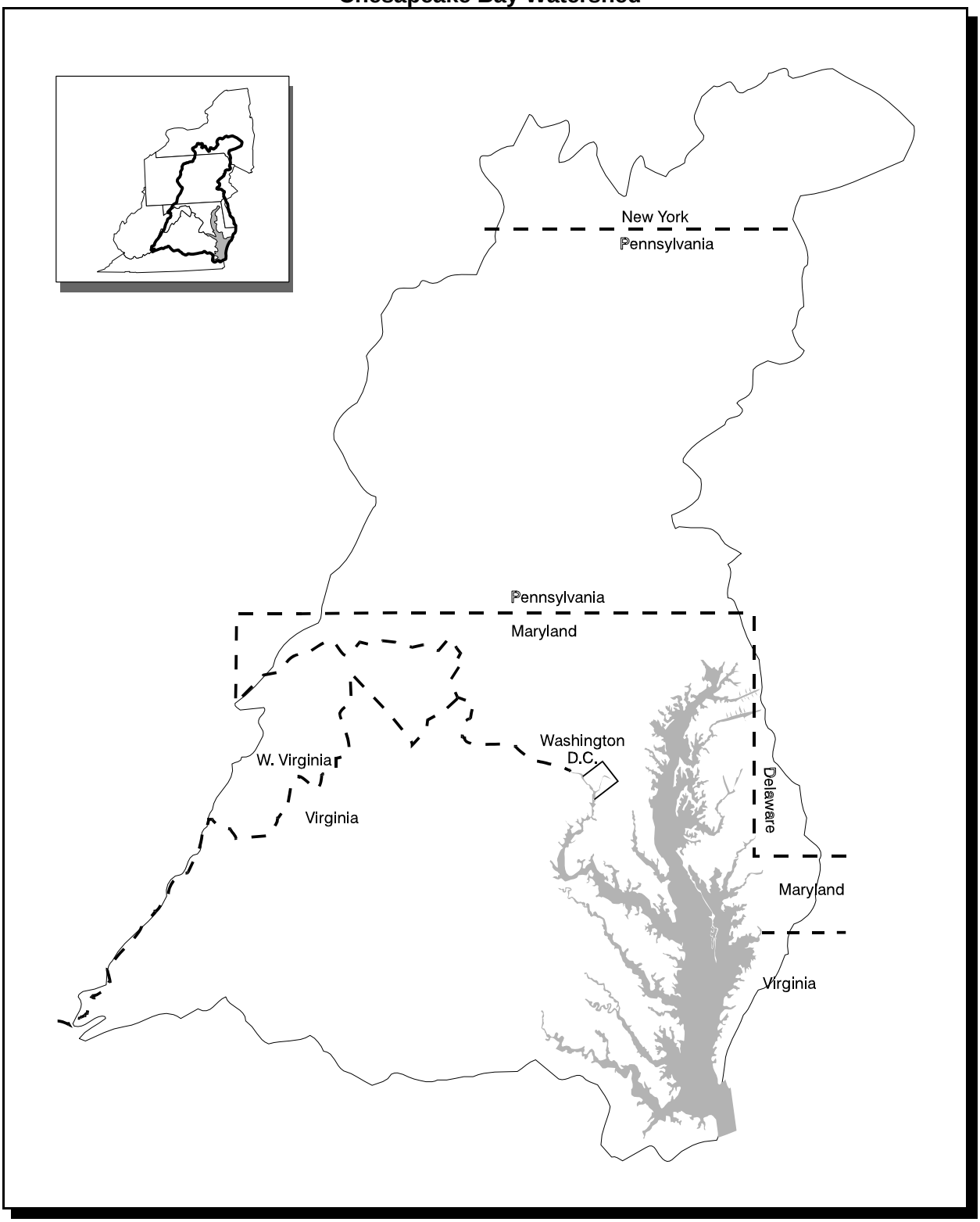
Supporting 295 species of finfish, 45 species of shellfish, and 27,000 plant species, the Chesapeake Bay is a national ecological treasure. The Chesapeake Bay is also home to 29 species of waterfowl and is a major resting ground along the Atlantic Migratory Bird Flyway. Every year, one million waterfowl winter in the Bay's basin. Economic highlights of the Chesapeake Bay are presented in the sidebar above. In all, the Chesapeake is a commercial and recreational resource for more than 14 million Bay basin residents.

The remainder of Section IV.C presents information on: the Chesapeake Bay Program; atmospheric deposition of nitrogen to the Chesapeake Bay; and atmospheric deposition of toxic contaminants to the Chesapeake Bay.

### *Chesapeake Bay Program*

Now in its fourteenth year, the Chesapeake Bay Program is a unique, regional partnership that has directed and coordinated Chesapeake Bay restoration since the signing of the historic 1983 Chesapeake Bay Agreement. The principal partners in the Chesapeake Bay Program include the State of Maryland, the Commonwealths of Virginia and Pennsylvania,

**FIGURE IV-8**
**Chesapeake Bay Watershed**



the District of Columbia, the Chesapeake Bay Commission (representing the state legislatures), and EPA on behalf of the federal government.

In 1983, EPA identified an excess of the nutrients nitrogen and phosphorus, and the resulting accelerated eutrophication, as the primary reason for the decline in water quality in Chesapeake Bay (U.S. EPA 1983). Excess nutrients stimulate "blooms" of phytoplankton algae, which then sink to the bottom of the Chesapeake. In the bottom waters, decay of the phytoplankton consumes oxygen, which expands the area of anoxic bottom waters (i.e., "dead waters" largely devoid of oxygen and unable to support life). Blooms of algae also reduce light to submerged aquatic vegetation (SAV), resulting in the loss of an important habitat for juvenile fish and crabs. (A recent assessment of the state of the Bay is presented in the sidebar.)

The state of the Chesapeake restoration and protection effort was described in the latest *State of the Chesapeake Bay* report (CBP 1995b):

"If the health of the Bay could be likened to that of a hospital patient, the doctor would report that the patient's vital signs, such as living resources, habitat, and water quality, are stabilized and the patient is out of intensive care. Some vital signs, such as striped bass and Bay grasses have improved dramatically, while a few, such as oysters, are in decline. Other vital signs are mixed but stable. Nutrients are being reduced, with phosphorus levels down considerably more than nitrogen levels and dissolved oxygen remains steady. Overall, the patient still suffers stress from an expanding population and changing land use, but it is on the road to recovery. Taken as a whole, the concentrated restoration and management effort begun ten years ago has produced tangible results--a state of the Bay that is better today than when we started..."

Using the watershed as the central focus, the Chesapeake Bay Agreement of 1983 recognized the historical decline of the Bay's living resources and recommended a cooperative approach among the federal and state governments within the watershed to address problems defined by the 1978-1983 Chesapeake Bay Research Program. The one-page agreement committed the signatories to work together to "fully address the extent, complexity, and sources of pollutants entering the Bay." The watershed approach of the state-federal partnership was chosen as the most practical method for implementing restoration efforts on both a local and regional scale.

Building on an expanded understanding of the Bay system and increasing experience with on-the-ground implementation within the cooperative basinwide partnership, a new Chesapeake Bay Agreement was signed in 1987 that set forth a comprehensive array of goals, objectives, and commitments to address living resources, water quality, growth, public information, and governance (Chesapeake Executive Council 1987). The centerpiece of the agreement was a commitment to achieve a 40 percent reduction of nitrogen and phosphorus entering the Bay by the year 2000. This measurable goal added a specific direction to ongoing monitoring, modeling, and nutrient reduction implementation programs. Through the 1987 Bay Agreement, the signatories also committed to "quantify the impacts and identify the sources of atmospheric inputs on the Bay system." This seemingly minor commitment at the time set the stage for a decade-long path to formally address atmospheric deposition as an integral component of basinwide pollution reduction strategies and implementation actions.

## *Atmospheric Deposition of Nitrogen to Chesapeake Bay*

This section presents information on the Chesapeake Bay Nutrient Reduction Strategy, an overview of atmospheric nitrogen loadings to the Bay (from modeling of the airshed, to nitrogen loadings estimates, to modeling of the watershed and estuary), and areas of uncertainty and work underway. Although the Chesapeake Bay Agreement and the baywide Nutrient Reduction Strategy focus on two main nutrients, nitrogen and phosphorus, this section focuses mainly on nitrogen because atmospheric deposition, the focus of this report, is a significant pathway of concern for nitrogen loadings only.

### NUTRIENT REDUCTION STRATEGY

The Chesapeake Bay Agreement commits the signatories to reduce the "controllable" nutrient loads by 40 percent by the year 2000. Controllable loads are defined as the baseline year loads minus the loads delivered to the Bay under an all- forested watershed (i.e., a watershed providing only natural, uncontrollable sources of nitrogen) within the Bay Agreement signatory jurisdictions (Linker et al. 1996). In other words, controllable loads are defined as everything over and above the total phosphorus or total nitrogen loads that would have come from an entirely forested watershed in the States of Pennsylvania, Maryland, and Virginia, and the District of Columbia, given existing rates of atmospheric deposition. In this definition, point source loads are considered entirely controllable. In addition, for the Bay Agreement, emissions of nitrogen compounds leading to atmospheric deposition are considered uncontrollable. Nonpoint sources may be controllable or uncontrollable.

**Sources of Nitrogen Entering the Bay**

Sources of the 170.8 million kilograms of nitrogen delivered annually to the Bay include:

- Point source water discharges (23% or 39.3 million kg), such as sewage treatment plants;
- Atmospheric deposition directly to tidal waters(9% or 15.4 million kg) and indirectly to tidal waters (18% or 30.2 million kg); and
- Other nonpoint sources (50% or 85.9 million kg), such as runoff from agriculture and urban areas.

To measure the goal of reducing controllable nutrient loads by 40 percent, the Chesapeake Bay Program established a 1985 baseline of nutrient loads. The 1985 baseline load was defined using 1985 point source loads and a 1984-1987 average load for nonpoint sources. The Chesapeake Bay Program chose the average load of the 1984-1987 period as the base to be representative of nonpoint source loads for all tributaries, because river flow and associated nonpoint source loads may vary depending on rainfall. Table IV-7 presents the 1985 base load and 40 percent reduction target for the major tributary basins of the Bay, and Figure IV-9 presents the locations of the tributary basins. After the year 2000, the tributary nutrient reduction targets (i.e., the 1985 base load minus the 40 percent reduction target) become nutrient caps that are not to be exceeded at any time in the future even in the face of continued population growth and development of the watershed.

In 1992, the basinwide reduction goal was reevaluated and allocated among the ten major tributary watershed basins. The state jurisdictions, with direct involvement of the public, then developed comprehensive tributary-specific nutrient reduction strategies within the individual watersheds. As part of the 1992 amendments to the Chesapeake Bay Agreement, the signatories

committed "to incorporate in the Nutrient Reduction Strategies an air deposition component which builds upon the 1990 Amendments to the federal Clean Air Act and explores additional implementation opportunities to further reduce airborne sources of nitrogen entering Chesapeake Bay and its tributaries" (Chesapeake Executive Council 1992).

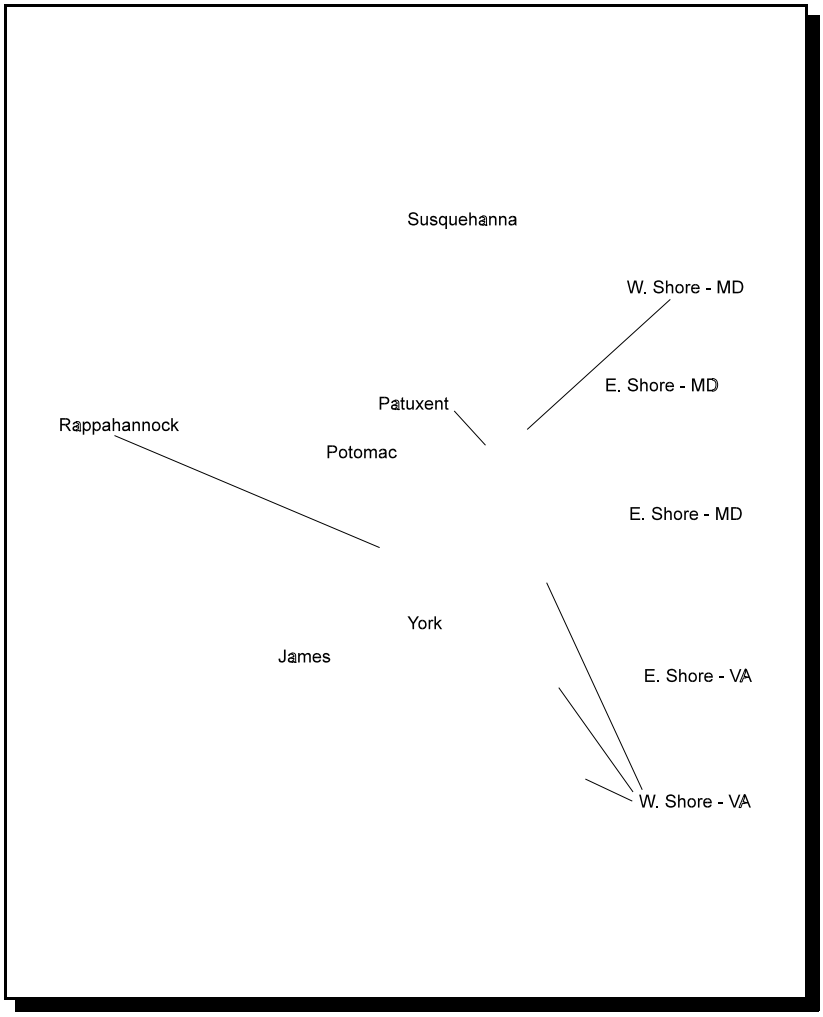
**TABLE IV-7**
**Chesapeake Bay Basin Nutrient Reduction and Loading Caps by Major Tributary Basin**
**(in millions of kilograms)**

| Major Tributary Watershed Basin | Nutrient | 1985 Base Load | 40% Target Reduction | Year 2000 Agreement Loading Cap |
|---|---|---|---|---|
| Eastern Shore MD | Nitrogen | 10.34 | 2.54 | 7.80 |
| | Phosphorus | 0.82 | 0.28 | 0.54 |
| Eastern Shore VA | Nitrogen | 0.82 | 0.18 | 0.64 |
| | Phosphorus | 0.04 | 0.01 | 0.03 |
| James[a] | Nitrogen | 19.82 | 6.39 | 13.43 |
| | Phosphorus | 2.80 | 0.97 | 1.83 |
| Patuxent | Nitrogen | 2.22 | 0.64 | 1.59 |
| | Phosphorus | 0.24 | 0.09 | 0.15 |
| Potomac[b] | Nitrogen | 31.16 | 8.48 | 22.68 |
| | Phosphorus | 2.41 | 0.78 | 1.64 |
| Rappahannock | Nitrogen | 3.76 | 1.18 | 2.59 |
| | Phosphorus | 0.39 | 0.15 | 0.24 |
| Susquehanna[c] | Nitrogen | 52.98 | 8.30 | 44.68 |
| | Phosphorus | 2.69 | 1.01 | 1.69 |
| York | Nitrogen | 2.90 | 0.86 | 2.04 |
| | Phosphorus | 0.42 | 0.15 | 0.27 |
| Western Shore MD | Nitrogen | 12.02 | 4.39 | 7.62 |
| | Phosphorus | 0.77 | 0.30 | 0.47 |
| Western Shore VA | Nitrogen | 1.91 | 0.54 | 1.36 |
| | Phosphorus | 0.23 | 0.09 | 0.14 |

[a] James loads include only loads from Virginia.
[b] Potomac loads include only loads from Pennsylvania, Maryland, Virginia, and the District of Columbia.
[c] Susquehanna loads include only loads from Pennsylvania and Maryland.

Source: Adapted from CBP 1992.

**FIGURE IV-9**
**Major Tributary Basins of the Chesapeake Bay**



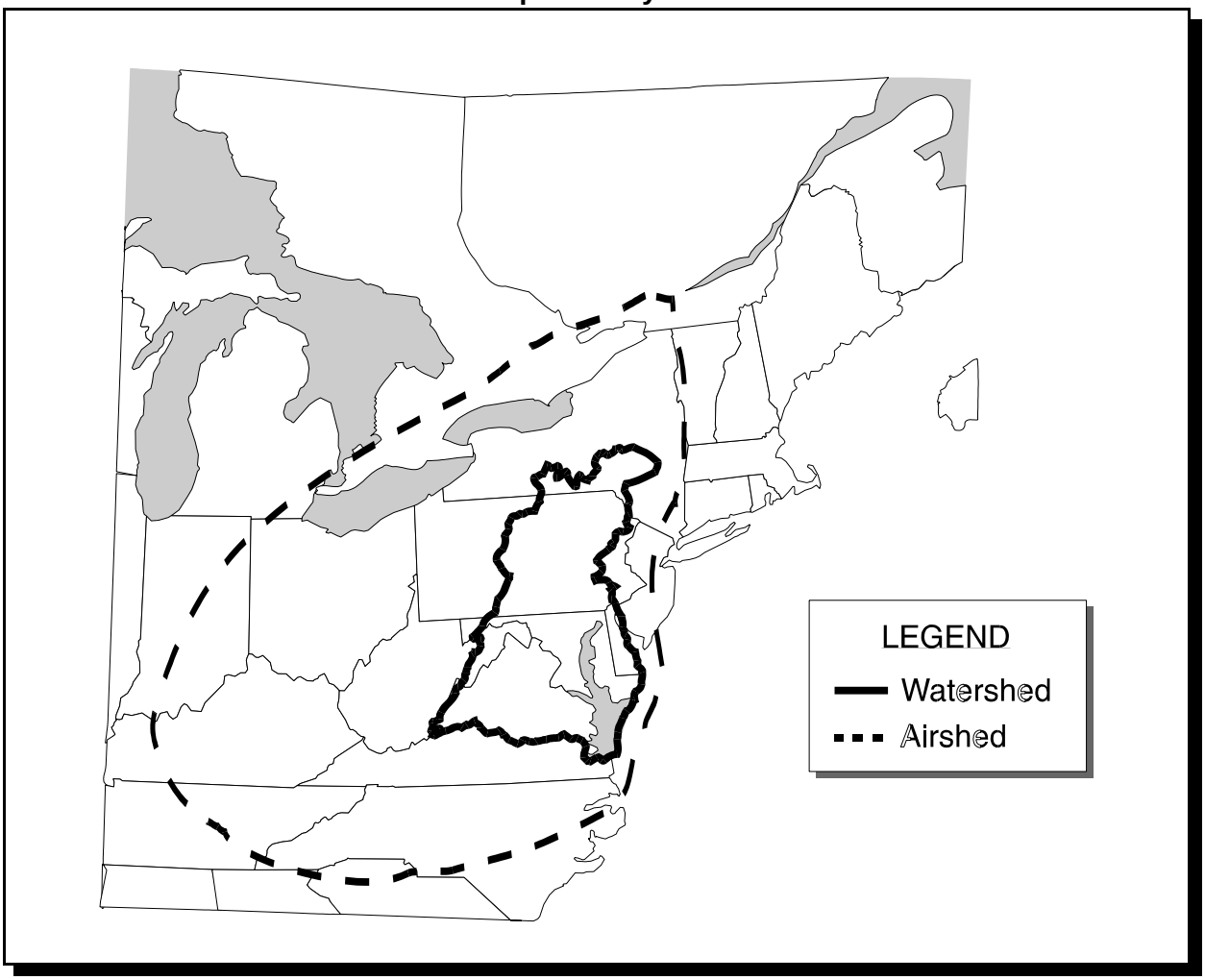

Any reductions in nitrogen loads brought about by programs implementing the CAA are considered to be additional nutrient load reductions separate from the point and nonpoint source reductions identified in the tributary nutrient reduction strategies. CAA implementation is expected to reduce nitrogen loads in Chesapeake Bay beyond the tributary strategy reductions (CBP 1994a). However, these additional reductions may last only a short time; at some time after the year 2000, population growth and increased land development are expected to begin eroding the gains made by the CAA. This expected increase in nutrient loads may make it difficult to meet the caps on nutrient loads to the Bay. Indeed, it was acknowledged at the time the tributary strategies were developed that "achieving a 40 percent nutrient reduction goal, in at least some cases, challenges the limits of current point and nonpoint source control technologies" (Chesapeake Executive Council 1992). To maintain the restoration progress that will be achieved by the year 2000, the technological limits of controls on reductions from point and nonpoint sources may have to be expanded to make further reductions in these areas economically attractive, or other sources of controllable nutrients may have to be considered to achieve cost-effective ecosystem protection in the Bay.

### MODELING AIR TO WATERSHED TRANSPORT: THE CHESAPEAKE BAY AIRSHED

A series of linked computer models have been developed by the Chesapeake Bay Program to simulate the transport of nitrogen from its emission sources to the Chesapeake Bay watershed and eventually into the tidal Bay waters. As a first step in establishing the air to tidal waters connection, the "airshed" of the Chesapeake Bay was defined. The boundaries of the airshed were defined as the contiguous areas whose sources "significantly" contributed (i.e., 75 percent) to atmospheric deposition of nitrogen to the Bay and its surrounding watershed (Dennis 1997). These boundaries were delineated by running a series of scenarios on the Regional Acid Deposition Model (RADM), using a predefined point of diminishing return (i.e.,

when a 50 percent reduction in emissions from large source regions would be expected to produce less than a 10 percent reduction in deposition onto the Bay watershed). The resulting 906,000 $km^2$ airshed, shown in Figure IV-10, is about 5.5 times larger than the Bay's watershed and includes: all of Maryland, Virginia, Pennsylvania, Delaware, the District of Columbia, West Virginia, and Ohio; most of New York; half of New Jersey, North Carolina, and Kentucky; and parts of Tennessee, South Carolina, Michigan, Ontario, and Quebec (including Lakes Erie and Ontario). (See Chapter III for a description of RADM and Dennis (1997) for more information on the use and limitations of RADM in this study.)

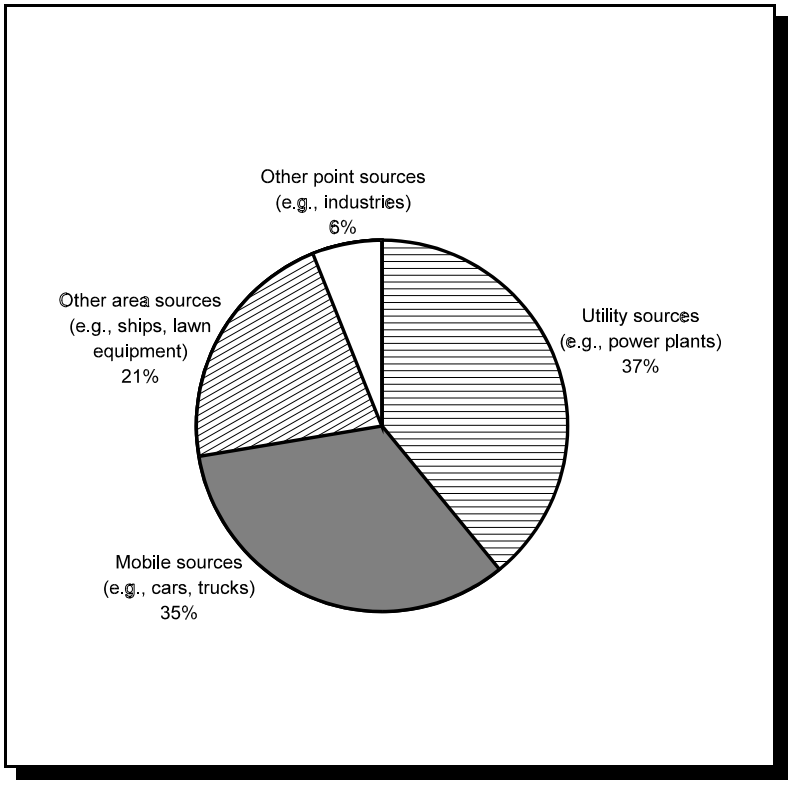
**FIGURE IV-10**
**Chesapeake Bay Airshed**




According to the Bay airshed model, about 25 percent of the nitrogen that is deposited on the Bay and its surrounding watershed originates from sources within the Bay watershed. Sources located within the jurisdictions of the Bay Agreement signatories of Maryland, Virginia, Pennsylvania, and the District of Columbia (including those sources that are within the state boundaries but outside of the Bay watershed) contribute about 40 percent of the nitrogen that deposits on the Bay and its watershed (Dennis 1997).

As defined, the Bay airshed, which accounts for 30 percent of all nitrogen emissions in the eastern United States and Canada, accounts for 75 percent of the atmospheric nitrogen deposited onto the Bay and its watershed. The remaining 25 percent of the deposition originates from emission sources outside the defined airshed (Dennis 1997). Therefore, the Chesapeake Bay airshed as defined here is smaller than the actual areas of the United States and Canada that contribute to nitrogen deposition to the Bay watershed. A still unresolved portion of the airshed is the portion that contributes to atmospheric deposition to offshore ocean waters which, in turn, contributes to the influx of nitrogen from coastal waters into the southern Chesapeake Bay (CBP 1994b).

**FIGURE IV-11**
**$NO_x$ Emission Sources in the Major Bay Influencing States**




Researchers compared results from the Bay airshed model to emissions inventory data on sources of $NO_x$ emissions and evaluated the contribution of these sources to nitrogen loads to the Bay. As shown in Figure IV-11, data from the emissions inventory indicate that the contributions from utility and mobile sources in the major Bay influencing states (i.e., Maryland, New Jersey, New York, Ohio, Pennsylvania, Virginia, West Virginia) to $NO_x$ emissions are roughly equal and make up the majority of emission sources. Through RADM, these data were confirmed and the patterns of nitrate deposition from the two sources were simulated. The model simulations suggest that utilities contribute a majority of the nitrate that deposits on the western side of the Bay watershed and that nitrate deposition from utility emissions shows a decreasing trend from the western to eastern portion of the watershed (see Figure IV-12[11]). These simulations further suggest that mobile sources, associated with $NO_x$ emissions from the Boston to Washington, D.C., metropolitan areas, contribute a majority of the nitrate that deposits along the Delmarva Peninsula, the Chesapeake Bay itself, and the lower portions of the western shore tidal tributaries (see Figure IV-13). In contrast to utility sources, the simulated deposition from mobile sources shows a decreasing trend from the eastern to western portion of the basin. Model scenarios simulating the effects of a uniform 50 percent reduction in nitrogen emissions from utilities alone and then from mobile sources alone show the same west to east, or east to west, gradients respectively (Dennis 1997).

[11] In Figures IV-12 and IV-13, a rough outline of the watershed and airshed is also shown. Each shaded area in these figures represents the percentage of all emissions that emissions from sources within the shaded area contribute to nitrogen oxides that deposit to the Bay.

**FIGURE IV-12**
**RADM Total (Wet and Dry) Nitrate Deposition from Utility Sources**
**(as a percent contribution of 1990 Base Case)**

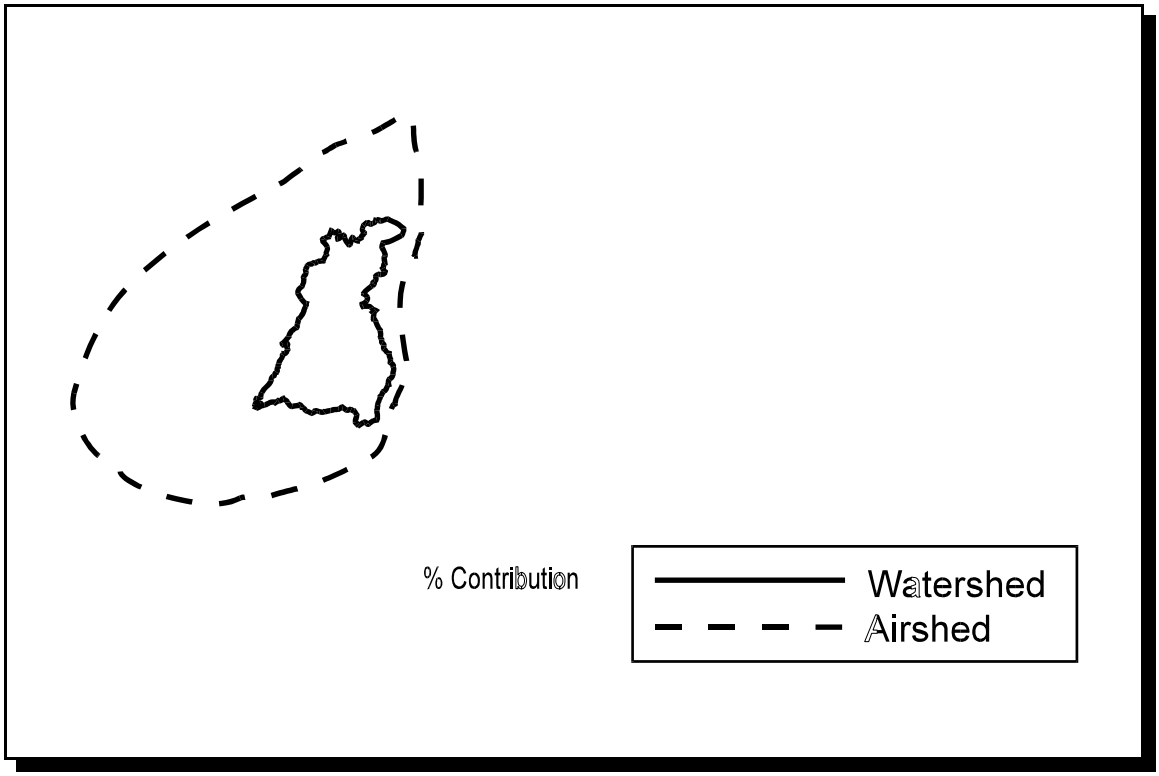
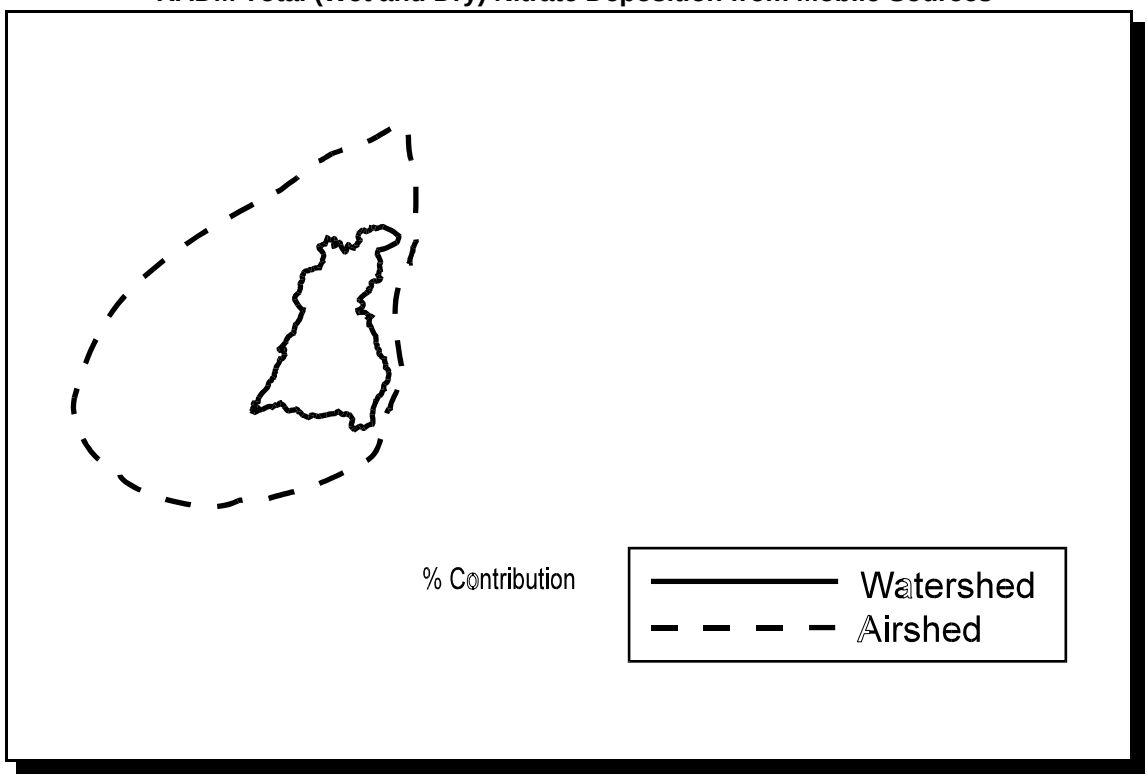



**FIGURE IV-13**
**RADM Total (Wet and Dry) Nitrate Deposition from Mobile Sources**



## ATMOSPHERIC NITROGEN LOADINGS TO CHESAPEAKE BAY

Atmospheric nitrogen loads from the airshed are transported to the Chesapeake Bay by three routes: direct deposition, both wet and dry, to the Bay tidal waters (i.e., direct loadings); indirect deposition, both wet and dry, to the watershed with subsequent runoff and river transport to the Bay (i.e,. indirect loadings); and deposition, both wet and dry, to adjacent offshore coastal waters with subsequent transport to the Bay through coastal currents. The first two processes, direct and indirect deposition to the Bay, are discussed below, as are some estimates of total loadings to the Bay using both a mass balance approach and computer models. The third pathway, deposition to offshore coastal waters, is the least understood route and is discussed later in this section under areas of uncertainty. Different nitrogen compounds that are measured or estimated in nitrogen loadings are discussed in the box on the next page.

**Direct Loadings**. The first estimates of atmospheric deposition to the tidal waters of Chesapeake Bay were made through spatial extrapolation of the National Atmospheric Deposition Program (NADP) sites in the Chesapeake watershed (Cerco and Cole 1994). The NADP is a long-term nationwide monitoring program that was started in the 1970s. Based on the annual loads reported by NADP, and an assumption that dry deposition of nitrate is equal to the long-term average of wet deposition of nitrate (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988), atmospheric deposition of inorganic nitrogen to the tidal waters of the Chesapeake was estimated as 6.4 million kilograms of nitrate and 1.8 million kilograms of ammonia. In addition, organic nitrogen was estimated as 6.8 million kilograms (Smullen et al. 1982). Studies have explored the idea that atmospheric deposition may contribute a significant proportion of phytoplankton nitrogen demands in coastal areas (Paerl 1985; Paerl 1988; Paerl et al. 1990); phytoplankton require nitrogen, both new and recycled, for growth. Fogel and Paerl (1991), for example, have estimated that 20 to 50 percent of annual new nitrogen demands for phytoplankton in Albemarle-Pamlico Sound, NC, may be supplied by direct atmospheric deposition to the water surface (wet and dry).

Using NADP data, wet deposition directly to the Chesapeake Bay tidal surface waters has been estimated to range from 3.3 to 4.2 million kilograms of nitrate per year (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988). Though NADP monitoring data allow initial estimates to be made of atmospheric deposition to the tidal Bay, it is not currently known if the over-land measurements of wet deposition accurately represent over-water wet deposition. To investigate this question, a daily precipitation chemistry site was established on Smith Island, Maryland, in late 1995. This site is providing the first time-series measurements of over-water wet deposition collected on the east coast.

Although the dry deposition to surface water loading rates of nitrogen compounds have been estimated for most nitrogen species over open ocean (Galloway 1985; Duce et al. 1991), these rates may not apply to coastal areas because of the different meteorological processes involved. Through the use of instrumented Chesapeake Bay Observing System (CBOS) buoys owned by the University of Maryland, estimates of nitrogen ($HNO_3$, $NO_2$, $NH_4$) dry deposition rates to the Bay tidal surface waters have been developed (Valigura 1995). These estimates corroborate those given by other investigators to some extent, but still cover a wide range of values, from 0.7 to 2.2 million kilograms of nitrate per year. From this data set, calculations were performed to determine the effect of atmospheric dry deposition on phytoplankton dynamics. This analysis demonstrated that dry deposited nitrogen may provide 10 percent of the annual "new nitrogen" demands by

phytoplankton in Chesapeake Bay, and that individual events could supply up to 75 percent of the new demands for periods of several days (Malone 1992; Owens et al. 1992).

### Characterization of Nitrogen Compounds

Most atmospheric nitrogen compounds (excluding $N_2$ and $N_2O$, which are relatively unreactive in the lower atmosphere) fall into two categories: reactive nitrogen and reduced nitrogen. In addition, some organic nitrogen species arise in the atmosphere from the interaction between nitrogen oxides and certain hydrocarbons. The relative portions of the different nitrogen forms in nitrogen loadings can vary widely based on source types and locations, proximity of sources to receiving waters, atmospheric transport, and physical and chemical transformations. Current estimates are that reactive nitrogen is the largest contributor to atmospheric deposition in coastal waters (40 to 60 percent), with ammonia (20 to 40 percent) and organic nitrogen (0 to 20 percent) also contributing significant amounts.

***Reactive nitrogen compounds***, primarily oxides of nitrogen, are emitted to the atmosphere through both natural and anthropogenic pathways, overwhelmingly (95 percent) as nitric oxide (NO). Natural NO sources include emissions from soils and generation by lightning; dominant anthropogenic sources include emissions from automobiles, power plants, and biomass burning. The dominant source of reactive nitrogen oxides present in air over North America is high-temperature combustion (e.g., power plants, automobiles). NO generated by combustion reacts quickly in the lower atmosphere, forming nitrogen dioxide ($NO_2$). The $NO_2$ is rapidly converted back to NO by ultraviolet light (photochemistry), then NO reacts again, resulting in a cycle driven by volatile organic compounds. From this photochemical cycle, ozone ($O_3$) is produced. The cycle is broken when $NO_2$ terminates into stable products, principally nitric acid vapor ($HNO_3$), and the NO gets used up. The ozone issue is therefore intimately related to the $NO_x$ (defined as NO + $NO_2$) question. $NO_2$ slowly deposits to the underlying surface (too slowly to break the cycle), but nitric acid vapor ($HNO_3$) is easily and quickly deposited. $HNO_3$ is the source of most of the reactive nitrogen deposited to the earth's surface.

***Reduced nitrogen compounds*** include ammonia ($NH_3$) and ammonium ($NH_4^+$). $NH_3$ is emitted into the atmosphere through both natural and anthropogenic pathways. Natural sources of $NH_3$ include microbial decomposition of organic nitrogen compounds in soils and ocean waters and volatilization from animal and human wastes. Anthropogenic sources include the manufacture and release of commercial and organic fertilizers during and after application and fossil fuel combustion. Human activities such as manure management and biomass burning exacerbate emissions from otherwise natural processes. $NH_3$ is a highly reactive compound and has a short residence time in the atmosphere. It is primarily emitted at ground level and quickly deposits to the area near its source unless it reacts with other gaseous chemicals (e.g., $SO_2$, $HNO_3$) and is converted to $NH_4^+$ aerosol (Asman 1994; Langland 1992). $NH_4^+$ can be transferred regionally as ammonium salts, such as ammonium nitrate $NH_4NO_3$ and ammonium sulfate $(NH_4)_2SO_4$, and these salts are the primary contributor to $NH_4^+$ concentrations measured in precipitation. Scavenging of $NH_3$ by precipitation can also be a major source of $NH_4^+$ in precipitation.

***Organic nitrogen*** may be a significant fraction of the total nitrogen measured in precipitation (Cornell et al. 1995; Gorzelska et al. 1997; Milne and Zika 1993). Data on the deposition of organic nitrogen has been limited, however, because of the paucity of reliable measurements, the historical variability in analytical techniques and results, and the current lack of suitable and uniform analytical measurement techniques. In fact, only wet deposition of dissolved organic nitrogen (DON) has been addressed. Various estimates for the relative flux of organic versus total nitrogen via wet deposition range from less than 10 percent to greater than 60 percent. The contribution of the unresolved organic fraction may significantly augment the atmospheric deposition of nitrogen to coastal waters. However, in addition to the lack of dry deposition data, there remain many conceptual questions related to source identification and the bioavailability of atmospheric organic nitrogen.

Sources: Luke and Valigura 1997; Paerl et al. 1997; Valigura et al. 1996.

**Indirect Loadings**. Quantifying indirect loadings of nitrogen to Chesapeake Bay, which refers to the atmospheric deposition of nitrogen to the terrestrial watershed and subsequent transport of the nitrogen from the terrestrial watershed to Bay surface waters, is an important component of the estimate of total atmospheric deposition of nitrogen to the Bay, yet it is largely uncertain. NADP monitoring data provide an initial estimate of the atmospheric deposition to the Chesapeake Bay watershed. Generally, higher deposition levels are found in the northern portions of the basin. In fact, some of the highest readings for atmospheric deposition of nitrate in the NADP monitoring network come from the northern sections of the Chesapeake basin. Greatest uncertainty is in dry deposition of nitrogen, which is not routinely measured by NADP.

The amount of atmospheric nitrogen transferred to surface waters within a given watershed depends on land use, total nitrogen loading from atmospheric, fertilizer, animal waste, and biosolid sources, the amount of soil nitrogen, characteristics of the soil, site rainfall and temperature, the elevation and slope of the land, and the type, age, and health of the vegetative cover. These characteristics vary independently, making it difficult to determine the fate of atmospherically deposited nitrogen over any area of significant size. However, several classification schemes for forested sites have been developed to evaluate a site's potential to retain and leach nitrogen (Melillo et al. 1989; Johnson and Lindberg 1992; Stoddard 1994).

One classification scheme in particular has helped organize thinking about nitrogen retention by classifying forest systems based on stages of nitrogen saturation (i.e., the extent to which the system is saturated with nitrogen; the more saturated a system, the more likely to leach nitrogen) (Stoddard 1994). The classifications range from Stage 0, where forest nitrogen transformations are dominated by plant and microbial assimilation (uptake) with little or no $NO_3$ export from the watershed during the growing season, to Stage 3, where nitrogen deposition is well in excess of assimilation and has reduced plant and microbial assimilation capacities resulting in greater export of atmospheric nitrogen as well as losses from mineralization of soil organic nitrogen. Study sites in the southern portions of the Chesapeake Bay watershed generally fall into the low nitrate export classification (Stages 0-1), while the northern portions have generally high to medium export classifications (Stages 1-2).

**Total Loadings Estimates Using A Mass Balance Approach**. The role of atmospheric transport as an important path for nitrogen deposition to estuarine areas was first publicized in 1988 (Fisher et al. 1988). Based on a mass balance analysis using 1984 hydrology data, the authors estimated that one-third of the nitrogen entering the Chesapeake Bay is deposited from the atmosphere. Several subsequent efforts (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988) to quantify atmospheric nitrogen loadings to Chesapeake Bay produced "best-estimate loadings" ranging from 25 percent (Fisher and Oppenheimer 1991) to about 33 percent (Hinga et al. 1991) of the total nitrogen loads to the Chesapeake. (A discussion of the uncertainties in a mass balance approach from one of these studies is presented in the sidebar on the next page.)

The approach taken in these mass balance studies can be divided into two components: (1) estimating wet and dry deposition; and (2) estimating nitrogen retention. A central difficulty in mass balance studies is the use of average land use values of nitrogen retention. Nitrogen retention assumptions used in three of the Chesapeake Bay studies are presented in Table IV-8.

**TABLE IV-8**
**Nitrogen Retention Assumptions Used in Chesapeake Bay Loading Studies**
**(as a percentage of nitrogen loading)**

| Land Use | Tyler 1988 | Fisher and Oppenheimer 1991[a] | Hinga et al. 1991[a] |
|---|---|---|---|
| Forest | 95.2-100.0 | 80.0 (51.0-100.0) | 80.0 (25.0-95.0) |
| Pasture | 93.7-99.96 | 70.0 (51.0-90.0) | 80.0 (25.0-95.0) |
| Cropland | 76.0-99.97 | 70.0 | 60.0 (45.0-75.0) |
| Residential | 62.0-95.3 | 35.0 (0.0-70.0) | 25.0 (10.0-50.0) |

[a] Numbers in parentheses indicate range tested.

Assembling an adequate understanding of long-term behavior when the processes involved are fundamentally episodic is another major challenge of contemporary models. Some studies indicate that the majority of the atmospheric wet deposition occurs during a few episodes (Dana and Slinn 1988; Fowler and Cape 1984), such that the wet-deposited nitrogen (as well as previously dry-deposited nitrogen) is deposited directly to, or flows quickly into, the surface waters without intermediate reduction in concentration. Despite these difficulties, mass balance studies provide a good first-order estimate of nitrogen loading to Chesapeake Bay.

Experimental manipulation at the watershed scale is being conducted at a few U.S. locations (Kahl et al. 1993; Norton et al. 1994). Work from these sites is providing information on the cycling of nitrogen in forested catchments and is fully supportive of the conclusion that atmospheric deposition contributes to nitrogen loading of coastal waters through the export of atmospherically derived nitrogen. Results of these long-term experiments are just beginning to be published. An example is the Bear Brook watershed in Maine. Divided into treatment and control catchments, the treated catchment received increased nitrogen loading in the form of labeled ammonia. The treated watershed response was an immediate increase in stream nitrate export (Norton et al. 1994; Uddameri et al. 1995).

**Inherent Uncertainties in Mass Balance Approach**

"It would not be difficult to make the [mass balance] calculations appear more elegant by subdividing the watersheds into more land use types, using a detailed data base of land uses, assembling more detailed lists of point source and agricultural inputs, and using some technique for contouring deposition over the watershed. None of these approaches are likely to make better calculations. More precise and reliable estimates of the magnitudes of inputs of atmospherically-deposited nitrogen to coastal waters will require significant advances in the understanding of many processes responsible for the behavior of nitrogen in terrestrial ecosystems and in rivers and streams."

Source: Hinga et al. 1991.

**Total Loadings Estimates Using the Chesapeake Watershed and Estuary Models**. The Chesapeake Watershed Model (discussed in more detail below) is one approach to disaggregating the separate components of terrestrial and river nitrogen dynamics in the basin, along with the temporal effects of high loading during rainfall events. The estimate of

atmospheric nitrogen loads for both direct and indirect deposition is 27 percent of the annual nitrogen load delivered to the Chesapeake Bay. This estimate was developed using the 1992 Watershed Model (Linker et al. 1993) and is consistent with the range reported by Chesapeake Bay mass balance studies (i.e., 25 to 33 percent). Further refinements are being made to the Watershed Model and an update of the estimate of atmospheric deposition is expected by September 1997.

To estimate wet deposition, the Chesapeake Bay Program combined output from a regression model developed from NADP weekly and daily precipitation chemistry measurements with data from the NOAA rainfall network. This approach yields daily estimates of rainfall to 74 sub-basins of the Chesapeake Bay watershed. Dry deposition was assumed to be equal to wet deposition for over-land areas and 44 percent of wet deposition for over-water areas. Indirect atmospheric loadings from the over-land portion of the watershed were estimated using the Chesapeake Bay Program Watershed Model.

The estimate of a 27 percent contribution of atmospheric deposition to total nitrogen loadings to the Chesapeake Bay falls within the range reported for other major eastern and Gulf coast estuaries, which are discussed in Section IV.D and summarized in Table IV-11 in that section.

#### MODELING THE CHESAPEAKE BAY'S WATERSHED AND ESTUARY

Water quality models of the Chesapeake Bay's watershed and estuary have been in use since the mid-1980s (CBP 1987; Donigian et al. 1991; Hartigan 1983; Thomann et al. 1994). The 1987 Bay Agreement's 40 percent nutrient reduction goal was based, in large part, on findings from these models.

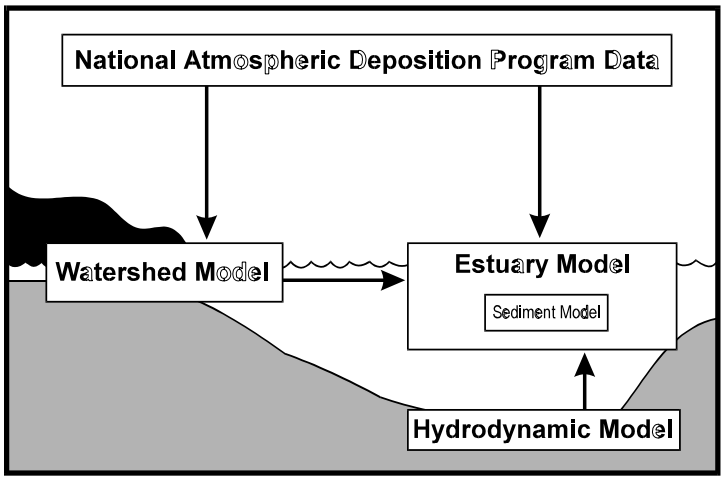
The first model of the Bay watershed was completed in 1982 and provided a basin-by-basin assessment of the relative importance of point source and nonpoint source controls of nutrients (NVPDC 1983). Subsequent refinements of the Watershed Model established the importance of animal waste management in the watershed (Donigian et al. 1991), the delivery to the Bay of atmospheric deposition loads from the watershed (Donigian et al. 1994), and the development of tributary allocation loads of nitrogen and phosphorus to achieve the 40 percent nutrient reduction goal (Thomann et al. 1994).

In a parallel effort, the first model of the Chesapeake estuary was completed in 1987 to evaluate the impact of nutrient reduction scenarios on the Bay's dissolved oxygen concentrations (CBP 1987). Results from the steady-state Estuary Model indicated that a 40 percent reduction in nutrient loads would significantly reduce anoxia (dissolved oxygen concentrations less than 1 mg/L) in the Bay mainstem during average summer (June-September) conditions (CBP 1988). The 40 percent controllable nutrient reduction goal, under the 1987 Bay Agreement, was based in large part on these findings.

A reevaluation of the Bay's nutrient reduction goal and a review of the progress made in reducing nutrients was scheduled for 1992. In advance of this reevaluation, researchers began refining and integrating the Watershed and Estuary Models (Figure IV-14). For example, the Watershed Model was updated with greater detail of agricultural and atmospheric sources and was linked to the Estuary Model (Donigian et al. 1994). Providing a predictive framework for

determining nutrient loads delivered to the tidal Bay under different source reduction scenarios, the Watershed Model simulated delivered nutrient loads with changes in land use practices and levels of wastewater treatment (Thomann et al. 1994). The Estuary Model was upgraded to add a sediment processes model and a hydrodynamic model, and was linked with the Watershed Model to accept Watershed Model nutrient loads as data input (Cerco and Cole 1994; DiToro and Fitzpatrick 1993; Johnson et al. 1991).

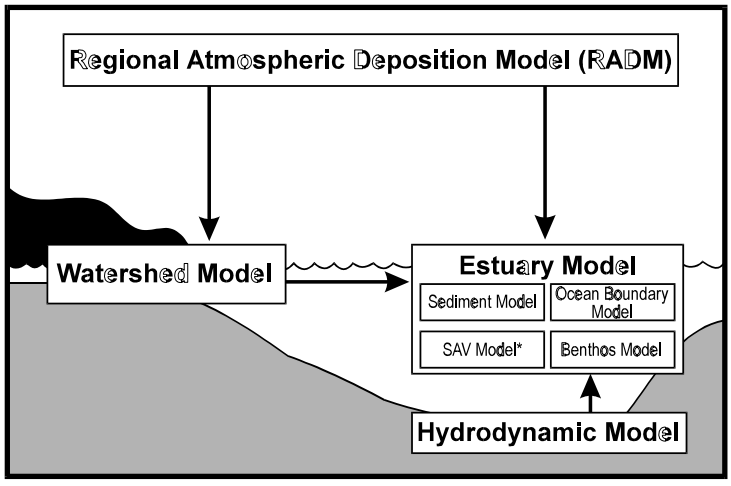
**FIGURE IV-14**
**Watershed and Estuary Model Integration**




The integrated Watershed and Estuary Model of the Chesapeake Bay was used to estimate water quality improvements that would be realized upon reaching the Bay Agreement goal of reducing controllable nutrients by 40 percent. Through the application of these models, the Bay Program partners established the Bay Agreement tributary nutrient allocations of nitrogen and phosphorus to be achieved by the year 2000 and maintained thereafter.

While this initial integrated model (Figure IV-14) could simulate the effects of atmospheric nitrogen deposition on water quality, it could not project the ultimate influence of changes in total nitrogen loadings (i.e., including loadings other than from the air) to the Bay. To provide for this predictive capacity, the Chesapeake Bay Program recently configured the Bay Watershed Model to accept daily atmospheric loadings by land use category (i.e., forest, pasture, cropland, and urban) (Linker and Thomann 1996). The Bay Watershed Model can now simulate the transport of increased or decreased atmospheric loadings to the Bay tidal waters along with nutrients from other land-based point and nonpoint sources. The Estuary Model is being upgraded to simulate basic ecosystem processes of submerged aquatic vegetation (SAV), benthic microorganisms, and major zooplankton groups. In addition, EPA's RADM is being directly linked to the Watershed and Estuary models. This new integrated model, functionally linking the airshed, watershed, estuary, and ecosystem, is expected to be completed in mid-1997. With these refinements, the

**FIGURE IV-15**
**Integrated Model Improvements**




* SAV = submerged aquatic vegetation

integrated model (Figure IV-15) will simulate and evaluate the overall loads of controllable and uncontrollable nitrogen from the surrounding airshed and watershed, and the impact of these loads on the ecosystem. This model will be one of the tools used by Chesapeake Bay Program state and federal managers to formulate additional nitrogen reduction steps needed to achieve the 40 percent reduction goal and maintain the cap on nutrient loadings after the year 2000.

**WATER QUALITY BENEFITS FROM REDUCING NITROGEN EMISSIONS**

Using the Watershed Model, several scenarios were developed to examine the effectiveness of air emission controls compared to traditional point source and nonpoint source controls on the delivery of nutrient loads to the Chesapeake Bay. The Watershed Model scenarios were:

- *Base Case Scenario*: This scenario represents the base year 1985 loads to the Chesapeake Bay.

- *Bay Agreement Scenario*: This scenario represents the 40 percent controllable nutrient load reduction to be achieved by the year 2000 (as discussed under Nutrient Reduction Strategy in this section).

- *Bay Agreement plus CAA Scenario*: The scenario represents the controls on point and nonpoint source loads through the Bay Agreement, plus the atmospheric load reductions expected under full implementation of the CAA titles I (reductions in ground level ozone), II (mobile sources), and IV (utility sources).

- *Bay Agreement plus Ozone Transport Commission (OTC) Scenario*: The scenario evaluates reductions from the controls on point and nonpoint source loads through the Bay Agreement, plus the effects of implementation of CAA titles I, II and IV, as well as additional nitrogen reductions to reduce ground level ozone in the mid-Atlantic and New England metropolitan regions as called for by the Ozone Transport Commission.

- *Limit of Technology Scenario*: This scenario estimates the nutrient reductions that may be realized using the current practical limit of point and nonpoint source control technologies, including conservation tillage for all cropland implemented; the Conservation Reserve program fully implemented; nutrient management, animal waste controls, and pasture stabilization systems implemented where needed; a 20 percent reduction in urban loads; and point source effluent controlled to a level of 0.075 mg/L total phosphorus and 3.0 mg/L total nitrogen. This scenario is significant because it determines the limit of currently feasible control measures.

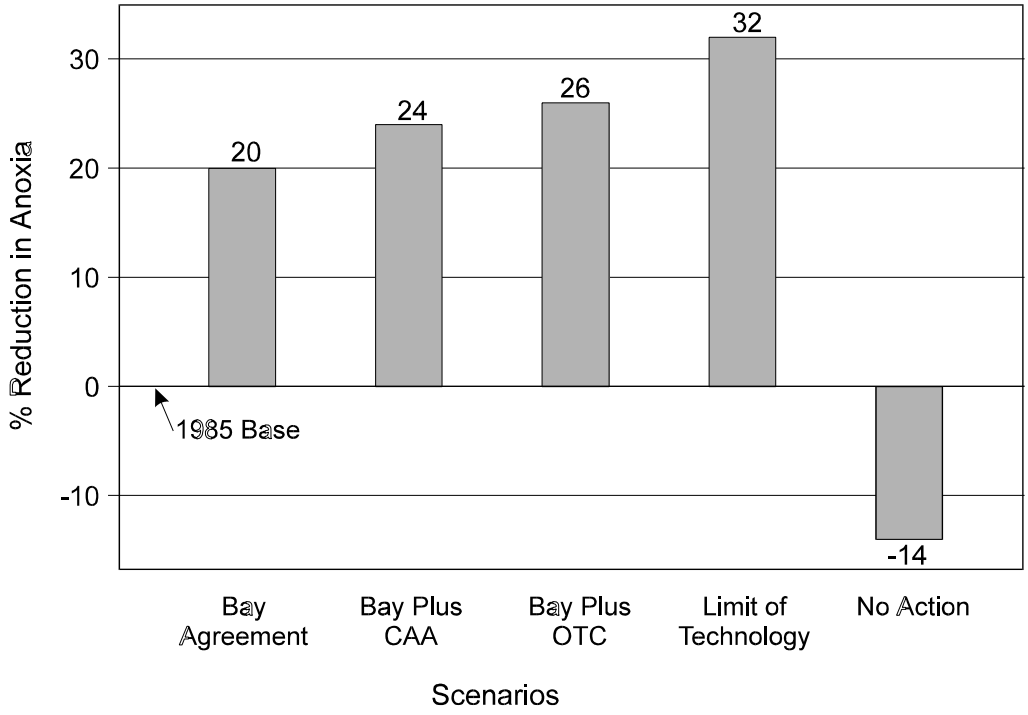
- *No Action Scenario*: This scenario represents loads that would occur in the year 2000 given population growth and projected changes in land use. The controls in place in 1985 were applied to the year 2000 point source flows and land use, representing the loading conditions without the nutrient reductions stipulated in the Bay Agreement.

These reduction scenarios are part of an effort to evaluate options for achieving the 40 percent nutrient reduction goal. Land-based nonpoint source and wastewater treatment facility-based point source reduction actions, planned for implementation in many Chesapeake tributary watersheds, are approaching the limits of technology. Options for reductions in air emissions are

being explored for maintaining the targeted 60 percent nutrient loadings cap beyond the year 2000 in the face of a growing population and resultant development in the watershed. Different options will have different costs and effectiveness with regard to water quality improvements, and a range of options should be evaluated to find the best approach.

The water quality improvement from the expected reduction in nitrogen emissions under each scenario are shown in Figure IV-16. The improvement in water quality reflects the estimated reductions in Bay bottom waters having no dissolved oxygen (i.e., reduction in Chesapeake anoxia or "dead waters"). Decreased nitrogen loadings will result in decreased water column nitrogen which will, in turn, decrease the growth of algae and improve the level of light penetration necessary to support the critically important SAV (Batiuk et al. 1992; Dennison et al. 1993; Thomann et al. 1994).

**FIGURE IV-16**
**Reductions in Anoxia Under Nutrient Reduction Scenarios**




The CAA and OTC scenarios indicate that these controls provide for nitrogen loading reductions and water quality improvements above and beyond those provided by implementation of the Chesapeake Bay Agreement commitment of 40 percent reduction in controllable nitrogen. Relative reductions from controls on sources of atmospheric deposition vary by tributary basin, with the more sensitive tributaries being the Susquehanna and the Potomac. These basins receive the highest deposition loads in the Chesapeake watershed and are among the most responsive to reductions in atmospheric deposition.

Though the differences between scenarios in percent reductions in anoxia might seem small, air emission controls could account for a fifth and a third of the baywide nitrogen load reduction goal through CAA implementation and OTC reductions, respectively. Such reductions could make achieving the 40 percent reduction target more feasible, and make maintaining a cap

on any further increases in nutrient loadings beyond 2000 possible. These additional reductions are especially important in the face of increasing population and watershed development that studies predict will increase the significance of atmospheric deposition as a source of nitrogen loadings to the Chesapeake Bay in the coming decades (Fisher et al. 1988; Pechan 1991).

### AREAS OF UNCERTAINTY AND WORK UNDERWAY

Nitrogen retention, the relative loadings of ammonia and organic nitrogen, and dry deposition to water surfaces are a few of the remaining areas of significant uncertainty in estimating atmospheric nitrogen loads. Several specific examples of areas of uncertainty are discussed below.

- ***Nitrogen retention*** within watersheds makes a big difference in the proportion of the atmospheric contribution to nitrogen loading to the Bay. Different retention assumptions in mass balance analyses lead to an uncertainty in the estimate of this contribution by more than a factor of two.

- ***Ammonia and organic nitrogen*** contribute a large portion of nitrogen deposition, perhaps more than 25 percent of the total atmospheric nitrogen load. However, it is unknown to what degree their sources are controllable, and they may be changing with time. For example, airborne ammonia emissions from agricultural animal operations could increase.

- Estimates of the relative contribution of ***dry deposition*** to the total atmospheric deposition loadings range from 25 to 63 percent (Duce et al. 1991; Levy and Moxim 1987; Logan 1983; Lovett and Lindberg 1986; Sirois and Barrie 1988; Walchek and Chang 1987). Faced with this wide range of estimates, many investigators choose to set the dry deposition loading equal to the measured wet deposition loading. This assumption is known to be questionable. While site-specific data to refine the estimate are lacking, recent evaluations indicate that dry deposition to tidal water surfaces is about 25 percent of wet deposition to tidal water surfaces (Luke and Valigura 1997).

- ***1990 baseline emission estimates*** continue to be refined. Estimates of emissions from off-road vehicles have been significantly improved. Ship emissions in harbors are suspected to be significantly underestimated (Booz-Allen & Hamilton 1991). While emissions from these sources are not large in the aggregate, they occur close to the Bay tidal surface waters, and thus have an influence greater than their national fractions would imply. Nitrogen emissions from on-road vehicles continue to be a source of uncertainty.

- ***Particulate nitrate*** (which has a low deposition velocity) and ***nitric acid*** (which has a high deposition velocity) are currently indistinguishable by RADM, leading to modeling uncertainty.

- The contribution of atmospheric nitrogen deposition to ***offshore ocean waters*** has not yet been characterized. The ocean waters exchange with waters of the Chesapeake Bay and thus may be a source, or a sink, of nitrogen loads to the Bay.

To reduce existing uncertainties in atmospheric loadings estimates, the Chesapeake Bay Program convened a workshop in June 1994, inviting scientists and managers with expertise and experience in understanding or managing atmospheric deposition. The challenge given to attendees was to construct a list of practical studies that would make the greatest impact on reducing the current uncertainty in estimates of the contribution of atmospheric deposition to declining aquatic ecosystem health. The resulting list (CBP 1995a) is summarized below:

- *Priority 1*: Conduct intensive, coordinated, and integrated monitoring studies at special locations within the watershed that characterize wet deposition, dry deposition, and local catchment area.

- *Priority 2*: Improve existing atmospheric models (e.g., reduce grid size, account for the effect of mountains).

- *Priority 3*: Improve models of chemical retention in watersheds.

- *Priority 4*: Improve emissions inventories and projections.

- *Priority 5*: Conduct measurements to extend vertical and spatial meteorological and chemical concentration coverage in models.

- *Priority 6*: Establish an extensive array of less intensive measurement stations to improve spatial resolution for selected variables.

To improve the cross-media modeling capabilities and to reduce existing sources of uncertainty in atmospheric deposition loadings estimates, the following work is underway through cooperation between EPA, state and federal agencies, and universities:

- Measuring the concentration of atmospheric organic nitrogen within the Chesapeake Bay watershed;

- Measuring dry deposition of nitrate to tidal surface waters of the Bay;

- Investigating the atmospheric deposition of dissolved organic nitrogen and its isotopic composition (delta (δ) $^{15}N$);

- Linking daily atmospheric deposition and resultant nitrogen runoff from pasture, forested, and urban lands within the Chesapeake Bay Watershed Model;

- Decreasing the grid size of RADM across the Bay watershed to increase the spatial resolution and improve the resultant model scenario output; and

- Linking RADM with the Watershed, Estuary, and Water Quality models, including simulation of atmospheric deposition to offshore ocean waters and exchange of the ocean waters with Chesapeake Bay waters.

The result of this and other work will become part of the integrated model of the Bay's airshed, watershed, estuary, and ecosystem (discussed above), which is expected to be completed in early

1997. A series of management scenarios, similar to the modeling scenarios discussed above, are also expected to be completed in 1997 to examine the most feasible and cost-effective combination of point source, nonpoint source, and air deposition reductions to meet the Chesapeake Bay Agreement commitment to cap nutrient loadings in 2000 at 60 percent of 1985 controllable base loadings and to ultimately restore the water quality conditions necessary to fully support the Bay's invaluable living resources.

## *Toxic Contaminant Deposition to the Chesapeake Bay*

### CHESAPEAKE BAY BASINWIDE TOXICS STRATEGY

The 1987 Chesapeake Bay Agreement committed the signatories to "develop, adopt, and begin implementation of a basinwide strategy to achieve a reduction of toxics consistent with the Clean Water Act of 1987, which will ensure protection of human health and living resources" (Chesapeake Executive Council 1987). The resultant strategy, adopted in 1989, initiated a multi-jurisdictional effort to more accurately define the nature, extent, and magnitude of Chesapeake Bay chemical contaminant problems and to initiate specific chemical contaminant reduction and prevention actions (Chesapeake Executive Council 1989). Building on a two-year reevaluation of the strategy and increased understanding of the nature of the Bay's toxics problems, a revised, farther-reaching strategy was adopted in 1994. The 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy* recognized the contribution of atmospheric deposition as a significant source of chemical contaminant loadings to the Bay. ***Within the basinwide strategy, the signatories committed to establishing a more comprehensive loadings baseline and setting an atmospheric deposition loading reduction target to be achieved over the next decade*** (Chesapeake Executive Council 1994).

In 1991, the Chesapeake Bay Program adopted its first Chesapeake Bay Toxics of Concern list, principally to identify and provide concise documentation on chemical contaminants that adversely affect the Bay or have a reasonable potential to do so. This list provided Chesapeake Bay region resource managers and regulators with a baywide consensus of priority chemicals and the information necessary to target these chemical contaminants for strengthened regulatory and prevention actions or additional research, monitoring, and assessment. Based on ambient concentrations of chemical contaminants and aquatic toxicity data, the toxic pollutants that represented immediate or potential threats to the Chesapeake Bay system were identified. The Toxics of Concern list (see sidebar) includes several pollutants of concern for atmospheric deposition to the Great Waters (cadmium, chlordane, lead, mercury, PAHs, and PCBs). In addition, a Chemicals of Potential Concern list was identified for the Chesapeake Bay (see sidebar) and includes two pollutants of concern for the Great Waters (dieldrin and toxaphene). Clear evidence is lacking that the contaminants on the Chesapeake Bay list of Chemicals of Potential Concern actually cause or have reasonable potential to cause adverse effects in the environment, but the Chesapeake Bay Program believes these

**Chesapeake Bay Toxics of Concern**

**Toxics of Concern**: atrazine, benz(a)anthracene,[a] benzo(a)pyrene,[a] cadmium, chlordane, chromium, chrysene,[a] copper, fluoranthene,[a] lead, mercury, napthalene,[a] polychlorinated biphenyls (PCBs), and tributyltin.

**Chemicals of Potential Concern**: alachlor, aldrin, arsenic, dieldrin, fenvalerate, metolachlor, permethrin, toxaphene, and zinc.

[a] A polycyclic aromatic hydrocarbon (PAH).

chemicals warrant enough concern to be carefully monitored and tracked. For example, a number of the chemicals listed as being a potential concern are either banned or restricted pesticides that have residues still remaining in the ecosystem at elevated levels but below thresholds of concern; others are chemicals of increasing concern due to use patterns or potential for toxicity to Bay resources.

In response to a commitment within the 1994 Basinwide Toxics Strategy, the Toxics of Concern list is currently being evaluated and revised using a risk-based chemical ranking system incorporating source, fate, and exposure/effects ranking factors. In-depth analyses of the top-ranked chemicals will lead to the selection of a revised Toxics of Concern list in 1997.

### CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC DEPOSITION STUDIES

Studies conducted in the southern Chesapeake Bay in the early 1980s suggest that the atmosphere is a significant source of organic contaminants to the Bay, such as organic carbon (Velinsky et al. 1986) and anthropogenic hydrocarbons, including PAHs (Webber 1993). While these earlier studies demonstrated the potential importance of the atmosphere in supplying contaminants to the Chesapeake Bay, they were limited by their methodologies (i.e., bulk deposition sampling, which is imprecise) and their relatively limited temporal and spatial scope. An assessment of the extent of toxic contamination in the Bay is presented in the sidebar.

**Extent of Toxic Contamination in Chesapeake Bay**

Prior to adoption of the 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy*, the Chesapeake Bay Program conducted a two-year, in-depth evaluation of the nature, extent, and magnitude of toxic contaminant-related problems within the Chesapeake Bay. Through the evaluation, no evidence was found of severe baywide impacts from chemical contamination, unlike other problems facing the Chesapeake Bay, such as the impacts from excess nutrients. The Program did document severe, *localized* toxicity problems, adverse effects from chemical contamination on aquatic organisms in areas previously considered unaffected, and widespread low levels of chemical contamination in all Bay habitats sampled.

Existing state and federal regulatory and management programs continue to reduce the input of potentially toxic chemicals to the Chesapeake Bay. Measured concentrations of many of these chemical contaminants in the Bay's bottom sediments, shellfish, fish, and wildlife have also generally declined, although elevated levels occur in several industrialized areas and some increasing trends have been observed. Progress in reducing the point sources of these chemical contaminants is offset by significant nonpoint source inputs of chemical contaminants (e.g., urban stormwater runoff, atmospheric deposition) from increasing development and urbanization of the Bay watershed.

To further explore the issue of atmospheric loadings of toxic contaminants to the Bay, the Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established by a team of scientists from the University of Maryland, Virginia Institute of Marine Sciences, University of Delaware, and Old Dominion University. The primary objective of the CBADS network was to provide the best estimate of total annual atmospheric loadings of a variety of trace elements and organic contaminants directly to the surface waters of the Chesapeake Bay. Because accurate estimates of baywide annual loadings require characterizing the spatial and temporal variability in contaminant loads in the atmosphere and in depositional fluxes to the Bay, CBADS collected data to help characterize this variability.

Based on previous studies of wet deposition in the region (e.g., Tyler 1988) and given the resources available for the network, three non-urban shoreline locations -- Wye Institute and Elms Institute, Maryland, and Haven Beach, Virginia -- were selected and sampled from 1990 to 1993. These three sampling site locations, as well as other monitoring sites around Chesapeake Bay, are shown in Figure IV-17. In establishing this initial network, the influence of urban areas was purposely avoided by locating the sites more than 50 kilometers from metropolitan areas (similar to the initial non-urban stations for the Great Lakes deposition network, the Integrated Atmospheric Deposition Network (IADN)). By minimizing possible urban influences, the resulting CBADS loading estimates are considered to be minimum values.

CBADS evaluated atmospheric loadings directly to the Bay only. Although it is most likely that some fraction of the toxic contaminants deposited from the atmosphere to the watershed of the Chesapeake Bay are ultimately transported to the surface waters, this study did not attempt to characterize indirect loadings for two main reasons:

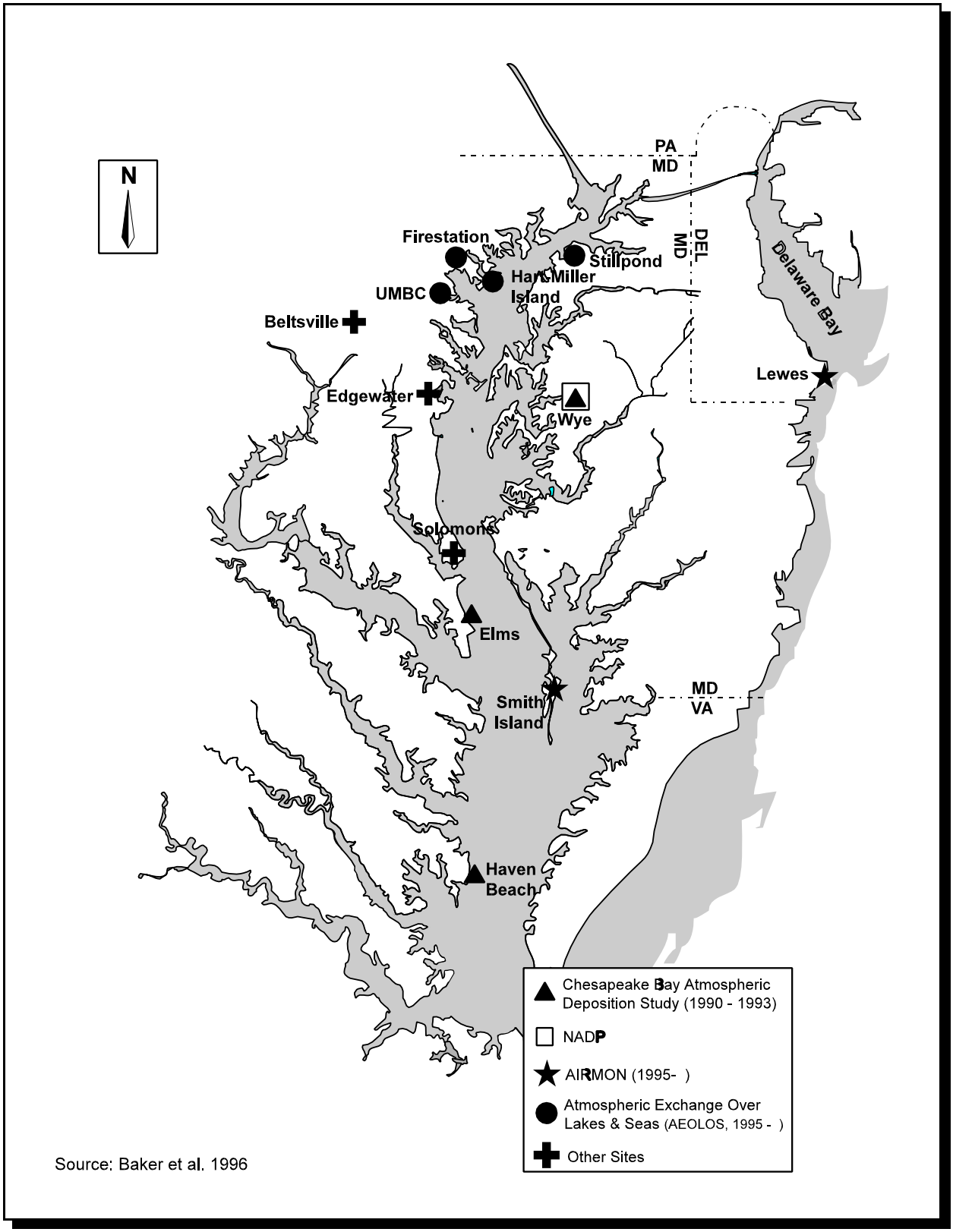
- Because deposition to the various land surfaces is likely much different than that to the water surface, fluxes measured at the shore-based stations cannot be extrapolated with confidence to the watershed; and

- The large uncertainty in the understanding of the fate of materials deposited to the land surface (i.e., the fraction transmitted to the receiving water) precludes the simple estimation of the indirect atmospheric loading of contaminants to the Chesapeake Bay in more than a rough estimate.

The CBADS data on concentrations in air, concentrations in precipitation, and wet and dry aerosol depositional fluxes are presented in Baker et al. (1997) and are summarized below. These data were collected for two groups of contaminants: trace metals (aluminum, arsenic, cadmium, chromium, copper, iron, manganese, nickel, lead, selenium, and zinc); and organic contaminants (14 different PAHs and total PCBs). Cadmium, lead, PAHs, and PCBs are pollutants of concern for atmospheric deposition to the Great Waters. Mercury data are being collected but have not yet been compiled, and data on other Great Waters pollutants of concern were not collected. For detailed results, a discussion of sampling methods, or a description of the limitations of the study, refer to Baker et al. (1997).

**Concentrations in Air**. Air concentrations for trace metals were determined by measuring the elemental composition of aerosol particles less than 10 $\mu$m in diameter. The elemental composition was dominated by the crustal elements aluminum and iron, as well as sulfur (in the form of sulfate). During 1991 and 1992, concentrations averaged over the three sampling sites were 116, 111, and 2,123 ng/m$^3$ for aluminum, iron, and sulfur, respectively. Trace element concentrations averaged over the same period ranged from 0.16 ng/m$^3$ for cadmium to 12.6 ng/m$^3$ for zinc, with lead averaging 3.88 ng/m$^3$. The fraction of each element derived from non-crustal (e.g., combustion) sources was estimated based on the average concentration of elements in the Earth's crust (Turekian and Wedepohl 1961), and assuming all of the measured aluminum associated with aerosol particles is derived from erosion of soils. In the Chesapeake Bay region, non-crustal sources supply greater than 95 percent of most of the elements measured on aerosol particles (Wu et al. 1994). Arsenic, cadmium, lead, sulfur, and selenium are almost exclusively non-crustal, and are likely introduced into the atmosphere as a result of combustion of fossil fuels and incineration of municipal wastes.

**FIGURE IV-17**
**Sampling Locations for**
**Chesapeake Bay Toxic Contaminant Atmospheric Deposition Studies**



The measured concentrations of trace elements were generally within a factor of two among the three sampling sites. On an average annual basis, concentrations slightly decrease from north (Wye) to south (Haven Beach), except sulfur, which is 15 percent higher at Elms (1991 and 1992) and Haven Beach (1992) than at the Wye site. The general decreasing trends observed from north to south, along with increasing sulfate, may indicate higher levels of low sulfur combustion sources (e.g., incinerators, vehicles) in the northern reaches of the Chesapeake Bay. In general, the spatial variability in the atmospheric concentrations of trace elements between sites (from north to south) is substantially lower than corresponding temporal trends.

Semivolatile organic chemicals exist in the atmosphere as gases and also are associated with aerosol particles (Pankow 1987). In this study, baywide annual average concentrations of PAHs in air ranged from 16 ng/m$^3$ for dibenz[*a,h*]anthracene to 2,590 ng/m$^3$ for phenanthrene. Atmospheric concentrations were quite variable with individual measurements ranging from one-tenth to ten times the annual average concentrations. These variations likely result from sampling air masses coming from differing directions, from changes in local and regional emissions, and from differences in atmospheric degradation and deposition rates. These variations show a seasonal pattern. For example, increased concentrations of gas-phase PAHs, such as pyrene, during the summer months may reflect both higher temperatures (i.e., enhanced volatilization) and increased coal and oil combustion to meet the electricity demand of air conditioning. Increases in the atmosphere of particulate PAHs, such as benzo[*a*]pyrene, may result from local burning of yard wastes and of wood for home heating. Some variation in atmospheric levels of organic chemicals may result from the efficient removal of particulate PAHs by precipitation (Poster and Baker 1996a, 1996b). In general, the magnitude of fluctuations in atmospheric levels of organic chemicals is larger than the corresponding variations in trace elements and sulfur discussed above, suggesting the importance of local combustion sources. Air concentration data were not available for PCBs.

**Concentrations in Precipitation**. The overall volume-weighted mean concentrations of trace elements in precipitation collected at the three sites range from 0.03 $\mu$g/L for cadmium at Elms to 14.6 $\mu$g/L for iron at Haven Beach. For lead, the range of overall volume-weighted mean concentrations was between 0.42 and 0.52 $\mu$g/L at the three sites. The relative proportion of trace elements in precipitation is nearly identical to that in the Chesapeake Bay aerosol particles, confirming that aerosol scavenging is the source of trace metals to wet deposition. Trace metal wet depositional fluxes are highly variable, changing more than ten-fold between consecutive months with little apparent seasonal dependence. This variability, which was similar at each of the three sites, results not only from fluctuations in the atmospheric inventories of trace metals, but also from changes in the amount of precipitation. On an annual basis, the volume-weighted mean concentrations of most trace metals did not systematically vary between the summer of 1990 and fall of 1993, again suggesting that these rural sites were most strongly influenced by the same regional background signal throughout the study.

While individual precipitation events result in spikes in trace metal deposition at one site that are not observed at the other two locations, these isolated events average out over the year. Annual volume-weighted mean concentrations of trace metals in precipitation are generally within a factor of two among the three sites, with no clear systematic spatial trend observed for all metals.

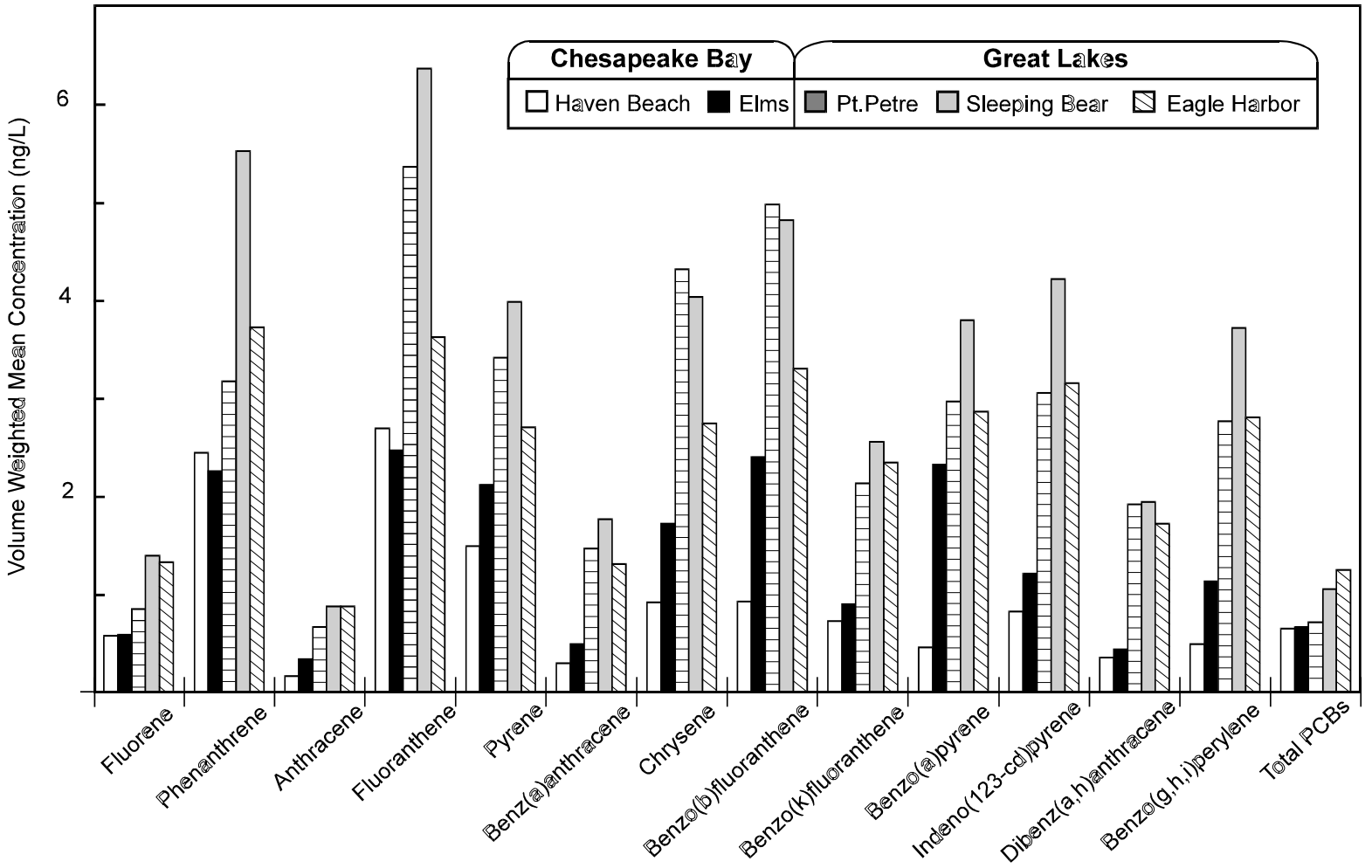
Overall volume-weighted mean concentrations of PAHs in precipitation ranged from 0.3 ng/L for anthracene and dibenz[*a,h*]anthracene at the Havens Beach site to 9 ng/L for pyrene at the Elms site. Volume-weighted mean concentrations for total PCBs were 1.1 and 0.9 ng/L at the Elms and Haven Beach sites (collected from June 1990 through September 1993), and 0.35 ng/L at the Wye site (collected from January to September 1993). As was observed for trace metals, wet depositional fluxes of organic contaminants varied considerably with time, and were dominated by episodic spikes at each location. Extremely high concentrations of some analytes, including pyrene, that were measured in both air and precipitation at the Elms site in the summer of 1990, may have resulted from local vegetation burning. Unlike PAH levels in the atmosphere, concentrations in precipitation did not systematically vary with season. The enrichment at the Elms site is especially pronounced for higher molecular weight PAHs, suggesting a local combustion source (e.g., wood burning for residential heating).

Concentrations of organic contaminants in precipitation measured in this study are consistently lower than those observed in the Great Lakes region (see Figure IV-18). For example, PAH concentrations in precipitation at the Chesapeake Bay sites are one-third to one-half as high as at the three IADN sites located at rural, shoreline sites on Lakes Ontario, Michigan, and Superior (Gatz et al. 1994). In contrast, levels of the same PAHs in the air over Chesapeake Bay are equal to or perhaps higher than those measured over the Great Lakes. Whether the apparent enrichment in PAHs in Great Lakes precipitation relative to that in the Chesapeake Bay region, as shown in Figure IV-18, is due to more efficient scavenging by precipitation in the colder, relatively drier Great Lakes region, or simply reflects methodological differences between the two networks, is unclear.

**Wet and Dry Aerosol Depositional Fluxes**. Using CBADS data, researchers calculated depositional fluxes (see Baker et al. (1997) for methodologies). Because these "depositional fluxes" are actually gross fluxes directly to the water surface and do not account for net gas exchange across the air-water interface, the term "deposition rates" is used in the remainder of this discussion in place of "depositional fluxes" to be consistent with the rest of the report in the use of the term "flux."

Total annual deposition rates in 1992 ranged from 0.07 $mg/m^2$ for cadmium at the Wye site to 121 $mg/m^2$ for aluminum at the Elms site; the highest annual deposition rate for lead was 1.34 $mg/m^2$ at the Wye site. Not surprisingly, dry aerosol deposition comprises the majority of the total deposition rate for the soil elements aluminum and iron, which occur on coarse particles. Wet deposition contributes between one-third and one-half of the total depositional rate of the remaining trace elements. Naturally, spatial trends in total deposition result from variation in precipitation chemistry and amount, and the trace element inventories associated with aerosol particles (given the considerable uncertainty in the dry aerosol deposition calculation, the same deposition velocity was used at each site). Although a distinct north to south trend in precipitation amount occurred in 1992 (100, 107, and 122 cm, respectively), total annual deposition rates were remarkably similar among the three stations. Total deposition rates were also very similar between years, again indicating that the relatively rare spikes in concentration are dampened against the chronic regional background signal.

**FIGURE IV-18**
**Comparison of 13 PAHs and Total PCBs in Precipitation (1992) from Chesapeake Bay and Great Lakes Sampling Sites**




Source: Baker et al. 1996 (Chesapeake Bay) and Gatz et al. 1994 (Great Lakes).

For PAHs, total annual deposition rates for 1992 range from 0.2 $\mu g/m^2$ for anthracene at the Wye site to 10.8 $\mu g/m^2$ for benzo[*b*]fluorathene at the Elms site.[12] Both wet deposition and dry aerosol deposition contribute to total PAH deposition, with dry aerosol deposition becoming relatively more important for the higher molecular weight, less volatile compounds. Total deposition rates for PAHs decrease with time during this study, with the lowest rates occurring during the first nine months of 1993. While some of this decrease is attributed to beginning with anomalously high measurement in the summer of 1990, decreases in both wet and dry aerosol deposition rates continued between 1992 and 1993. The total annual deposition rate for total PCBs is about 3.5 $\mu g/m^2$, with approximately equal contribution from wet and dry aerosol deposition.[13]

Overall, total annual deposition rates for PAHs and PCBs are generally within 50 percent among the sites. Given the uncertainty in the dry aerosol deposition estimates, this percentage indicates little spatial variability when integrating over annual cycles. However, this study did not specifically address the possible influences of urban areas, such as the cities of Baltimore, Washington, and Norfolk, on atmospheric deposition, which may be important.

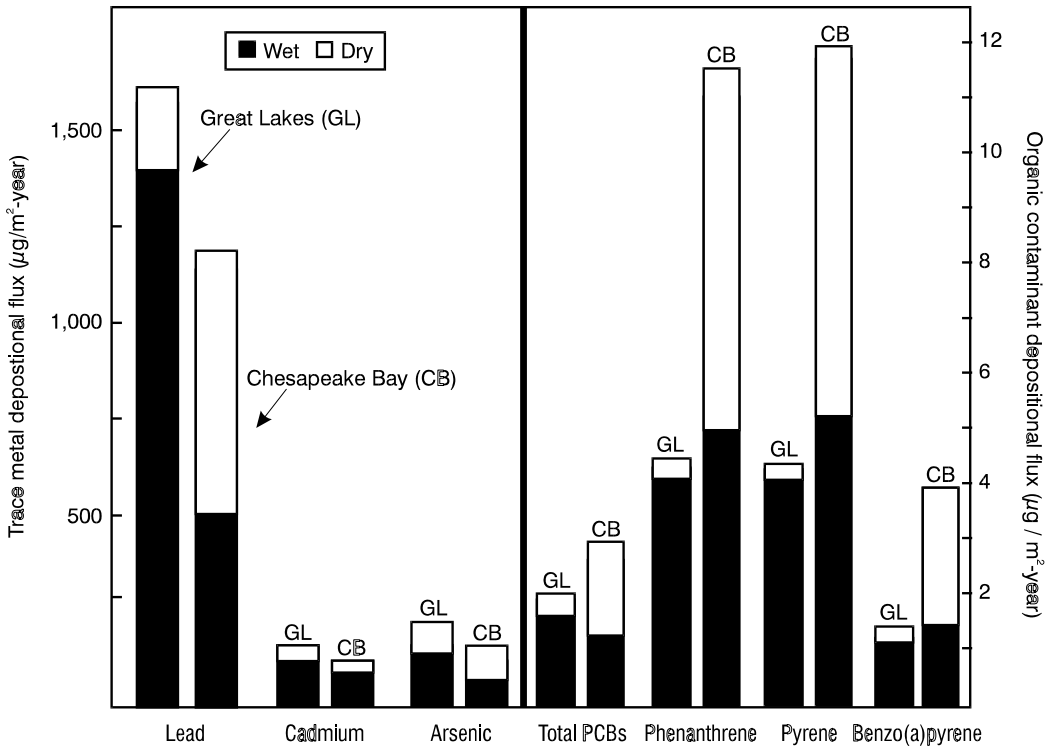
To place the atmospheric deposition rates calculated in this study in perspective, they are compared to similar estimates made for the Great Lakes region (Figure IV-19). Wet deposition rates for lead and arsenic are almost three times higher in the Great Lakes than in Chesapeake Bay, despite significantly less rainfall (80 versus 110 cm/year); wet deposition rates for cadmium are similar for both regions. Wet deposition rates for PAHs and total PCBs are fairly similar between the two regions, as higher concentrations in Great Lakes precipitation (see Figure IV-18) are offset by lower precipitation amounts. Dry aerosol deposition rate estimates are higher in the Chesapeake region, especially for organic contaminants.[14] In addition, measured aerosol-bound organic concentrations were generally higher than the values used in the Great Lakes dry aerosol deposition calculations (Eisenreich and Strachan 1992). Despite the differences, estimated atmospheric deposition rates are generally within a factor of two between the Chesapeake Bay and the Great Lakes regions, which, given the numerous opportunities for error in these measurements and calculations, is quite good agreement.

[12] Because the "fluxes" in this study are actually gross fluxes directly to the water surface, these data do not take into account exchange of gaseous organic contaminants across the air-water interface. Other recent studies have shown that this is the *dominant* atmospheric deposition process for semi-volatile organic contaminants, including PCBs (Baker and Eisenreich 1990; Achman et al. 1993) and low molecular weight PAHs (Nelson et al. 1995). In those studies, the net direction of exchange is often from the water to the air and diffusive gas exchange is large enough to offset wet deposition and dry aerosol deposition.

[13] Because aerosol particle-associated PCBs were present below analytical detection limits, estimates of PCB dry deposition were made using an aerosol PCB level calculated from the measured gaseous PCB concentration and the Junge-Pankow sorption model (Pankow 1987; Leister and Baker 1994).

[14] This is due, in part, to the choice of deposition velocities used for the two studies (0.2 cm/sec for all species in the Great Lakes, 0.26 cm/sec for trace elements and 0.49 cm/sec for organics in the Chesapeake Bay). All of these values are within the generally accepted range for dry aerosol deposition velocities.

**FIGURE IV-19**
**Comparison of Chesapeake Bay and Great Lakes Atmospheric Depositional Fluxes**




Source: Baker et al. 1996 (Chesapeake Bay) and Eisenreich and Strachan 1992 (Great Lakes).

### CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC LOADINGS

To estimate the annual baywide loadings of trace elements and organic contaminants to the Bay, the annual site-specific wet and dry aerosol deposition rates were averaged and the two average rates were multiplied by the surface area of the Bay. Uncertainties in these loading estimates are likely, on the order of a factor of two, mainly due to the inability to estimate dry aerosol loadings on a finer temporal resolution.

Baywide atmospheric loadings of aluminum and iron are estimated at 1,340,000 and 799,000 kg/year, respectively (see Table IV-9). Loadings of trace elements range from 1,110

kg/year for cadmium to 49,400 kg/year for nickel; lead loadings are estimated at 12,500 kg/year. Loading estimates are generally similar for 1991 and 1992, except for nickel and zinc due to

**TABLE IV-9**
**Annual Atmospheric Loadings of Trace Metals and Organic Contaminants to the Chesapeake Bay[a]**

| Pollutant | Wet Deposition (kg/year) | Dry Deposition (kg/year) | Total Deposition (kg/year) |
|---|---|---|---|
| Aluminum[b] | 137,000 | 1,200,000 | 1,340,000 |
| Arsenic | 607 | 1,050 | 1,660 |
| Cadmium | 867 | 240 | 1,110 |
| Chromium | 1,026 | 2,030 | 3,060 |
| Copper | 5,575 | 3,620 | 9,200 |
| Iron | 132,800 | 666,000 | 799,000 |
| Manganese | 13,200 | 13,600 | 26,800 |
| Nickel | 7,185 | 6,160 | 13,300 |
| Lead | 5,440 | 7,080 | 12,500 |
| Selenium | 1,390 | 2,930 | 4,320 |
| Zinc | 26,000 | 22,800 | 49,400 |
| Total PCBs | 13 | 20 | 37 |
| PAHs | | | |
| Anthracene | 6 | 6 | 13 |
| Benz(*a*)anthracene | 9 | 34 | 44 |
| Benzo[*a*]pyrene | 17 | 36 | 53 |
| Benzo[*b*]fluoranthene | 36 | 98 | 134 |
| Benzo[*e*]pyrene | 21 | 67 | 88 |
| Benzo[*ghi*]perylene | 19 | 75 | 94 |
| Benzo[*k*]fluoranthene | 22 | 65 | 88 |
| Chrysene | 29 | 85 | 114 |
| Dibenz[*ah*]anthracene | 7 | 16 | 22 |
| Fluoranthene | 70 | 119 | 189 |
| Fluorene | 16 | 12 | 27 |
| Indeno[123*cd*]pyrene | 20 | 78 | 98 |
| Phenanthrene | 63 | 92 | 155 |
| Pyrene | 75 | 109 | 184 |

[a] To estimate annual baywide loadings of elements and organic contaminants to the entire Chesapeake Bay, the annual site-specific wet and dry aerosol fluxes were averaged and these two average fluxes were multiplied by the surface area of the Bay.
[b] Contribution of aluminum is considered to be entirely from natural sources (i.e., not emitted through human activities).

Source: Baker et al. 1997.

elevated wet deposition measured at the Haven Beach site in 1992. Loadings of PAHs range from 13 kg/year for anthracene to 189 kg/year for fluoranthene. Total PCB loadings are estimated to be 37 kg/year. Interestingly, for many of the pollutants in Table IV-10, wet deposition and dry aerosol depositional fluxes appear to decrease between 1991 and 1992. Whether this reflects a real inter-annual variation or simply results from aggregating measurements from different locations is unclear.

To place these loadings in perspective, they are compared in Table IV-10 to recent estimates of trace metal and organic contaminant loadings delivered to the Chesapeake Bay by the Susquehanna River (Conko 1995; Foster 1995; Godfrey et al. 1995). The Susquehanna River is the largest tributary of the Chesapeake Bay, supplying approximately 60 percent of the freshwater inflow to the estuary. Annual riverine loadings of dissolved and particulate trace metals and organic contaminants were determined by analyzing flow-weighted samples collected at Conowingo, Maryland, between February 1994 and January 1995 (Conko 1995; Foster 1995). Atmospheric deposition directly to the surface waters of the Chesapeake Bay supplies PAH loads that are comparable to or greater than the loads of dissolved PAHs delivered by the Susquehanna River (Table IV-10). Particulate-bound organic contaminants discharged from the river dominate the loading of PAHs, with a large contribution from the high sediment burden carried by the river during high flows. Dissolved total PCB loads from the river are approximately three times those from the atmosphere. Atmospheric depositional fluxes of several elements, including lead, cadmium, and chromium, are within a factor of two of the dissolved load from the Susquehanna River. Again, particulate metal loads from the river dominate over both dissolved riverine loads and atmospheric deposition.

While it is interesting to compare the relative importance of riverine and atmospheric sources of trace elements and organic contaminants to the Chesapeake Bay, the results should be carefully interpreted. While the Susquehanna River delivers large quantities of these substances to the Bay, much of this load is removed in the northern extreme of the Bay (Helz and Huggett 1987) and is delivered episodically during high river flows. Whether particulate-bound metals and organic contaminants are broken down in forms that can be taken up by aquatic organisms is quite unclear. In contrast, atmospheric deposition directly to the water's surface supplies these toxics directly to the water column, without any comparable zone of efficient removal. However, it has recently been suggested that combustion-derived PAHs associated with aerosol particles washed into the surface waters by precipitation also may not be broken down (McGroddy and Farrington 1995). Finally, the distinction between riverine and atmospheric loadings is not clear. Some fraction of the pollutant input from the tributaries results from deposition of atmospheric pollutants to the watershed, with subsequent transmission through the vegetation and soil layers into surface waters (i.e., indirect loading); however, this input cannot yet be quantified.

### AREAS OF UNCERTAINTY AND WORK UNDERWAY

Building on the existing CAA requirements, the Chesapeake Bay Program's state and federal partners will focus their efforts on implementation of the Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy commitment to "establish a more complete baseline and source identification for atmospheric deposition...and set a reduction target from that baseline to be achieved over the next decade" (Chesapeake Executive Council 1994). However, there are several remaining areas of uncertainty to be addressed related to atmospheric deposition of toxic contaminants to Chesapeake Bay. Two significant ones are:

**TABLE IV-10**
**Relative Importance of Sources of Trace Metals and Organic Contaminants to Chesapeake Bay**

| Pollutant | Susquehanna River Load (kg/year)[a] | | Atmospheric Deposition Load (kg/year)[b] |
|---|---|---|---|
| | Dissolved | Particulate | |
| Aluminum | 2,560,000 | 64,800,000 | 1,340,000 |
| Arsenic | 12,600 | ND | 1,660 |
| Cadmium | 2,130 | 26,700 | 1,110 |
| Chromium | 4,130 | 111,000 | 3,060 |
| Copper | 47,800 | 151,000 | 9,200 |
| Iron | 4,100,000 | 40,000,000 | 799,000 |
| Manganese | 3,290,000 | 1,530,000 | 26,800 |
| Nickel | 121,000 | 65,200 | 13,300 |
| Lead | 6,530 | 38,600 | 12,500 |
| Zinc | 77,900 | 360,000 | 49,400 |
| Total PCBs | 97 | 68 | 37 |
| PAHs | | | |
| Benz[*a*]anthracene | 12 | 364 | 44 |
| Benzo[*a*]pyrene | 5 | 436 | 53 |
| Chrysene | 15 | 316 | 114 |
| Fluoranthene | 108 | 1,020 | 189 |
| Fluorene | 37 | 85 | 27 |
| Phenanthrene | 63 | 388 | 155 |
| Pyrene | 104 | 925 | 184 |

[a] Annual loads entering the Chesapeake Bay via the Susquehanna River, measured at Conowingo, Maryland, between February 1994 and January 1995 by Foster (1995) for organic compounds and Conko (1995) for metals.
[b] Total atmospheric deposition loads directly to the surface of the Chesapeake Bay as measured by CBADS.

Source: Baker et al. 1997.

- *Dry Deposition*. Dry deposition is viewed as an important mechanism by which chemical contaminants are deposited onto the Bay's tidal surface waters and surrounding watershed. As is the case with nitrogen, there are no widely accepted techniques for direct measurement of dry deposition fluxes of metal or organic contaminants. Although no direct measurements of dry deposition directly to the Bay exist, depositional fluxes have been estimated based on a particle-size-dependent deposition velocity function applied to direct measurements of aerosol concentrations of metals and organic chemical contaminants. Given the absence of direct measures of dry deposition fluxes, there is much uncertainty associated with these loading estimates.

- *Transport through the Watershed*. Atmospheric deposition of a pollutant can be a direct input to the Bay surface waters or can be transported from the watershed by surface water and groundwater to the Bay. Transported loads are a component of the total fluvial (i.e., surface water) input from the watershed to the Bay. The degree of landscape retention for a given substance is related to the geomorphology, land use, basic hydrological characteristics unique for each watershed, and soil chemistry. Limited

studies to date suggest that the degree of watershed throughput is relatively small (< 30 percent of the rate of rainfall volume). However, evidence to date suggests that watersheds serve as a "reservoir" for atmospherically deposited metals; organically bound metals are sequestered but may be episodically mobilized by acidic precipitation. Because of the relatively large watershed to open water surface area ratio typical of coastal plain estuaries such as Chesapeake Bay (15:1), recent estimates for nitrogen and trace elements suggest that the indirect atmospheric loading may be as significant as the direct input. Thus, while it has been possible to quantify direct atmospheric flux with a fair degree of confidence, one of the primary uncertainties associated with resolving the total atmospheric loading to Chesapeake Bay is in gauging the indirect loading as it relates to the watershed transmission/retention for the myriad of sub-basins (Valigura et al. 1995).

To further improve existing estimates of the relative atmospheric deposition contribution to total chemical contaminant loadings to Chesapeake Bay, the following work is underway. In 1993, the University of Delaware, in cooperation with the U.S. Geological Survey, initiated a pilot study to investigate the transport of atmospherically deposited trace elements through a pristine, forested watershed in the headwaters of the Potomac River (Bear Branch). This study, funded by the Maryland Department of Natural Resources' Power Plant Research Program, has the following specific objectives: (1) to accurately determine the wet and dry atmospheric trace element loads into the watershed, (2) to compare the total atmospheric load versus fluvial output of trace metals and (3) to estimate the transport of atmospherically deposited trace elements through the watershed relative to the trace metals naturally relapsed during weathering of the soil and rock within the study area. The Bear Branch watershed was chosen as it has been well-characterized hydrologically, is representative of the land use in the Potomac basin (60 percent of which is forested), and possesses an unreactive quartzite lithology which simplifies its geochemical weathering behavior. Further watershed transmission studies began in the spring of 1996 in the Appalachian Plateau of Western Maryland. While the results of these studies will represent an initial attempt to quantify the watershed retention/transport of atmospheric loads, further work is needed to extend the study to other regions with differing land use/geomorphology, in order to accurately determine an integrated, baywide watershed transport factor.

The next section describes programs in other coastal waters, as well as research relevant to atmospheric deposition in these coastal waters.

## IV.D Coastal Waters

As stated previously, section 112(m) designates "coastal waters" as EPA's National Estuary Program (NEP) and NOAA's National Estuarine Research Reserve System (NERRS) estuaries. These two programs and EPA's Gulf of Mexico Program are the three significant coastal waters programs, outside the Chesapeake Bay Program, established in the last decade. Although the Gulf is not designated by name under section 112(m) of the CAA, 11 estuaries in various locations spanning the Gulf coastline are either NEP or NERRS (current or proposed) designated sites and, thus, are designated Great Waters.

The NEP, NERRS, and Gulf of Mexico programs differ in purpose and procedure, but they all serve to protect and restore the nation's valuable coastal water resources. The remainder of this section provides background information on each of these programs, followed by a discussion of studies of atmospheric deposition to coastal waters and future research needs.

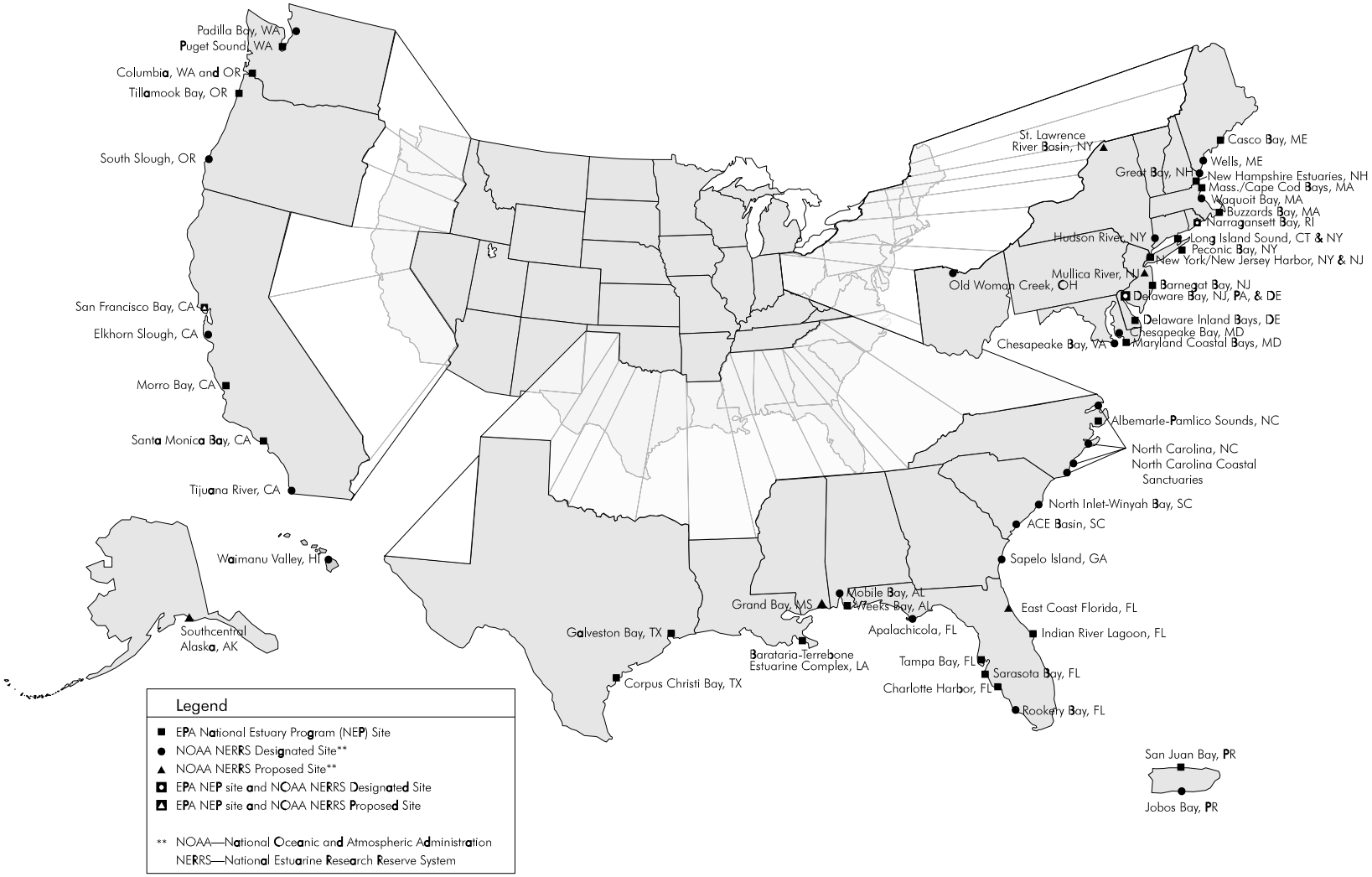
### *National Estuary Program*

Congress established the National Estuary Program (NEP) in 1987 under section 320 of the Clean Water Act. Through the NEP, states nominate estuaries of national significance that are threatened or impaired by pollution, development, or overuse. EPA evaluates the nominations and selects those estuaries for which there is evidence of political support, citizen and government involvement (local, state, regional, federal), and available scientific and technical information to address the identified problems. For the selected estuaries, EPA convenes management conferences with representatives from all concerned groups (e.g., industry, agriculture, environmental organizations, state agencies) to more fully characterize problems and seek solutions through a collaborative decision-making process. Through these conferences, Comprehensive Conservation and Management Plans (CCMPs) are developed, for which EPA provides up to 75 percent of the funding. Each management conference must complete development of the CCMP within three to five years of the date the conference was convened. Upon approval of the CCMP, action plans are carried out by implementation agencies involved with development of the plan.

The purpose of the NEP is to identify nationally significant estuaries, protect and improve their water quality, and enhance their living resources. The NEP currently includes 28 estuaries (individually called NEPs) representing a wide spectrum of environmental conditions (see Figure IV-20). Because there are over 150 estuaries in the United States and only a small fraction can be targeted for action through the NEP, the NEP is intended to act as a national demonstration program, such that results and lessons learned in the NEP estuaries are shared and applied by parties concerned with other estuaries throughout the country. It should be emphasized that the NEP is a management program rather than a research program and relies on the research of other agencies and institutions to support its work. The development of support networks and cooperation between local, state, regional, and federal agencies is one of the program's greatest assets.

Several NEPs have identified atmospheric deposition of pollutants as a concern to the health of their estuaries. These NEPs have either initiated studies on the contribution of atmospheric deposition to annual loadings for nitrogen and/or other pollutants or expressed serious interest to EPA in conducting such projects. Nine NEPs submitted proposals to EPA in early 1996 for funding under section 112(m) of the CAA. To date, only two NEPs (Tampa

**FIGURE IV-20**
**Locations of NEP and NERRS Sites**



Bay and Galveston Bay) have received funding under section 112(m) to conduct such work. Other NEPs (Casco Bay, Delaware Bay, Long Island Sound, Massachusetts Bays, and Peconic Bay) have initiated exploratory studies funded by their own program and other sources. Atmospheric deposition research related to the NEPs is discussed later in this section.

### *National Estuarine Research Reserve System*

Another program established to recognize the importance of estuaries is the National Estuarine Research Reserve System (NERRS), which was created by Congress in 1972 under the Coastal Zone Management Act and which operates under the authority of NOAA. The mission of NERRS is to establish and manage, through the cooperation of federal, state, and community efforts, a national system of estuarine research reserves that are representative of various regions and estuary types in the United States, in order to provide opportunities for long-term research, education, and stewardship.

The process for designating and maintaining a NERRS site includes five main activities, all of which may be partially funded by NOAA: (1) predesignation phase (includes selection of the site by the state and, after approval of the site by NOAA, preparation of a draft and final management plan and environmental impact study and completion of basic characterization studies); (2) acquisition of land and development activities; (3) after designation as an NERRS site, implementation of research, educational, and research programs detailed in the research reserve management plan; (4) estuarine research and monitoring; and (5) educational and interpretive activities.

Currently, 22 areas are designated as NERRS sites, including portions of Chesapeake Bay and associated lands in Maryland and Virginia (see Section IV.C for a detailed discussion of Chesapeake Bay). Six additional NERRS sites have been proposed or are in the beginning stages of development. See Figure IV-20 for the location of the NERRS estuaries.

Studies on the direct contribution of atmospheric deposition to NERRS waters are limited at this time. Available information on atmospheric deposition research related to NERRS waters is presented later in this section. For example, the indirect contribution of atmospherically deposited nitrogen to Waquoit Bay, Massachusetts, through its watershed has been estimated and modeled as part of the multi-year Waquoit Bay Ecological Risk Assessment Case Study (NOAA and MA DEM 1996; U.S. EPA 1996f).

### *Gulf of Mexico Program*

The Gulf of Mexico is a very important resource to all of North America. Its surface area is about 1,603,000 $km^2$, large enough to cover one-fifth of the continental United States. The U.S. portion of the Gulf's shoreline measures over 2,500 km from the Florida Keys to the Rio Grande. Taking into account the shoreline length of all the bays, estuaries and other coastal features of the Gulf, its effective shoreline length is about ten times that amount. The 21 major estuaries along the Gulf coast account for 24 percent of all estuarine area in the 48 contiguous states, and 55 percent of the marshes. The watershed of the Gulf includes more than two-thirds of the continental United States (plus one-half of Mexico and parts of Canada, Guatemala, and Cuba), with the Mississippi River watershed alone draining about 40 percent of the continental United States.

The Gulf of Mexico Program (GMP) was established in 1988 in response to citizens' concerns over declines in the Gulf's fish, shellfish, and wildlife; the quality of life in many coastal communities; the need to protect the remaining valuable resources and prevent problems before they occur; and to forge a positive relationship between ecological health and economic vitality of the Gulf region. The GMP is a unique organization that involves representatives from government agencies (federal, state, and local), business and industry, non-profit organizations and educational institutions, and interested individuals in the process of setting environmental goals and implementing actions to achieve those goals. The aim is to foster coordination and cooperation among these organizations in order to reduce costs and coordinate actions.

The GMP is not a regulatory program, but rather an approach to environmental protection, similar to the Chesapeake Bay Program, that is founded on the principles of:

- Partnership among government agencies, private, and non-government interests to define and characterize concerns and implement solutions;
- Sound science and information as the basis of informed decision-making to guide actions; and
- Public involvement to determine goals, identify solutions, and generate the consensus needed for action.

Since its beginning, the GMP has made significant progress in effectively involving a broad spectrum of the public in defining goals and objectives and in characterizing fundamental issues. The fundamental goals of the GMP are to:

- Protect human health and the food supply;
- Maintain and improve Gulf habitats that support living resources (fish, shellfish, and wildlife); and
- Maintain and enhance the sustainability of the Gulf's living resources.

In the past few decades, the Gulf of Mexico has been degraded, largely due to nutrient enrichment and habitat loss. The contribution of atmospheric nitrogen to nutrient enrichment is not well understood and is possibly a significant concern. Fed by nutrient-enriched waters of the Mississippi River, a large area of near-bottom waters commonly become depleted in oxygen, or hypoxic. At its peak, this area (known as the "hypoxic zone") can extend over a 18,192 $km^2$ area from the coastal waters of the Mississippi River Delta of Louisiana to those of eastern Texas. Stresses to the benthic (bottom-dwelling) community have been observed in this zone, including mortality of larger non-swimming benthic organisms. This and other possible disruptions to the food chain threaten to affect the commercial and recreational fish species within the hypoxic area. In addition to the Louisiana Shelf hypoxic zone, 18 other coastal areas in the Gulf have experienced hypoxia due to increasing nutrient concentrations or loads. Evaluation of atmospheric deposition of pollutants to the Gulf is discussed below, including research in NEPs located in the Gulf, as well as two studies conducted in the Gulf as a whole.

## *Studies of Atmospheric Deposition in NEP and Other Coastal Waters*

Municipal and industrial wastewater discharges and urban runoff/storm sewer inputs have historically been considered the largest sources of pollutants to coastal waters. Recently, however, researchers have begun to investigate the role of atmospheric deposition as a source of pollutants in a coastal waters (e.g., Paerl 1985, 1993; Scudlark and Church 1996). Assessing the impact of atmospheric deposition of pollutants has become a priority for many NEPs and other coastal watershed protection programs. There is a clear need to characterize the types, quantities, and sources of pollutants that are being directly and indirectly deposited from the atmosphere into these estuaries. Recent studies on atmospheric deposition to coastal waters are discussed briefly below and are presented in Tables IV-12 and IV-13. Data as of December 1995 on the contribution of atmospheric deposition to nitrogen loadings to Chesapeake Bay and other coastal waters are presented in Table IV-11; in this table, information is presented first for Chesapeake Bay and a few related tributaries, followed by other coastal waters in descending order of tidal water area. Data on the contribution of atmospheric deposition to the loadings of toxic pollutants in coastal waters are presented in Table IV-12; the coastal waters are listed in geographical order clockwise from the northeast coast to the northwest coast. In general, the studies discussed below have evaluated the relative contribution of nitrogen and other pollutants of concern, and do not attempt to identify the particular emission sources contributing to this pollution.

**Nitrogen Loadings to Coastal Waters**

At least 40 studies around the world, the majority of which have been published since 1990, have addressed at least the direct loading component of atmospheric nitrogen loadings. However, the measurement and modeling techniques used vary considerably among individual studies, making comparisons difficult. Table IV-11 presents a summary of selected studies performed along the East and Gulf coasts of the United States that are comparable in broad terms. The two criteria for selecting these studies were that the study results were either published in a credible peer-reviewed journal or advocated by a major management organization (e.g., an EPA NEP). These studies can be divided into two groups: those that considered both direct and indirect nitrogen loads and those that considered only direct loads. Data from these studies show that, in general, the amount of atmospheric nitrogen input is related to the size of the waterbody and its watershed. To some extent, the percent load from atmospheric deposition is influenced by whether both direct and indirect deposition were considered.

As mentioned above, research on atmospheric deposition to Tampa Bay, Florida, and Galveston Bay, Texas, has been conducted under the Great Waters program. The Tampa Bay and Galveston Bay studies are discussed first, followed by a description of other studies of atmospheric deposition to NEP estuaries (i.e., those that have been funded through sources other than EPA's Great Waters program). Initial observations from these studies suggest that direct and indirect loadings from air deposition may be significant sources of nitrogen and toxic pollutants to coastal waters.

**Tampa Bay**. As recently as 1991, atmospheric deposition of nitrogen, air toxics, and other pollutants was assumed to have a minimal effect on water quality in Tampa Bay. However, based on a methodology developed by the Environmental Defense Fund (Fisher et al. 1988), early calculations provided an early indication of likely nitrogen loadings from atmospheric deposition in Tampa and Galveston Bays.

## TABLE IV-11
## Estimates of Atmospheric Nitrogen Loadings to Selected Coastal Waters[a]
## (in millions of kg)

| Coastal Water | Surface Area ($km^2$) | | Deposition to Watershed | Direct Deposition to Tidal Waters | Indirect Atmos. Load From Watershed | Total Atmospheric Load | Total Load From All Sources | % Load from Atmosphere | Reference[b] |
|---|---|---|---|---|---|---|---|---|---|
| | Watershed | Tidal Waters | | | | | | | |
| Chesapeake Bay (MD/VA) | 165,886 | 11,400 | 175 | 16 | 29 | 45 | 170 | 27 | 5 |
| Rhode River (MD) | 33 | 4.9 | — | 0.005 | — | 0.005 | 0.012 | 40 | 6 |
| Choptank River (MD) | 1,779 | 361 | — | 0.57 | — | 0.57 | 1.54 | 37 | 11 |
| Patuxent River (MD) | 2,393 | 137 | — | 0.22 | — | 0.22 | 12.6 | 13 | 11 |
| Potomac River (MD) | 29,940 | 1,210 | — | 1.9 | — | 1.9 | 35.5 | 5 | 11 |
| New York Bight (NY/NJ)[c] | 50,107 | 38,900 | 69 | 54 | 8 | 62 | 164 | 38 | 1 |
| Albemarle-Pamlico Sound (NC) | 59,197 | 7,754 | ~39 | 3.3 | 6.7 | 10 | 23 | 44 | 4 |
| Long Island Sound (NY/CT) | 43,481 | 4,820 | 43 | 5 | 6 | 11 | 60 | 20 | 3 |
| Massachusetts Bays (MA) | — | 3,700 | — | 1.6-6 | — | 1.6-6 | 22-30 | 5-27 | 15 |
| Delaware Bay (DE) | 36,905 | 1,846 | 53 | 3 | 5 | 8 | 54 | 15 | 2 |
| Tampa Bay (FL) | 6,216 | 1,031 | — | 1.1 | — | 1.1 | 3.8 | 28 | 14 |
| Guadalupe Estuary (TX) | — | 551 | — | 0.31 | — | 0.31 | 4.2-15.9 | 2-8 | 13 |
| Narragansett Bay (RI) | 4,708 | 328 | 4.2 | 0.3 | 0.3 | 0.6 | 5 | 12 | 1 |
| | | | — | 0.4 | — | 0.4 | 9 | 4 | 12 |
| Newport River Coastal Waters (NC) | 340 | 225-1,600 | — | 0.095-0.68 | — | 0.095-0.68 | 0.27-0.85 | 36-80 | 4 |
| Sarasota Bay (FL) | 524 | 135 | — | 0.16 | — | 0.16 | 0.6 | 26 | 10 |
| Delaware Inland Bays (DE) | 800 | 83 | — | 0.28 | — | 0.28 | 1.3 | 21 | 9 |
| Flanders Bay (NY) | 83 | 39 | — | 0.027 | — | 0.027 | 0.36 | 7 | 8 |
| Waquoit Bay (MA) | ~70 | ~8 | 0.062 | — | 0.0065 | 0.0065 | 0.022 | 29 | 7 |

[a] Estimates as of December 1995.

[b] (1) Hinga et al. 1991; (2) Scudlark and Church 1993; (3) Long Island Sound Study; (4) Paerl and Fogel 1994; (5) Linker et al. 1993; (6) Correll and Ford 1982; (7) Valiela et al. 1996; (8) Peconic Bay NEP; (9) Delaware Bays NEP; (10) Sarasota Bay NEP 1995; (11) Boynton et al. 1995; (12) Nixon et al. 1995; (13) Brock et al 1995; (14) Tampa Bay NEP, Zarbock et al. 1994; (15) Massachusetts Bays NEP 1996.

[c] New York Bight extends from Cape May, New Jersey, to Long Island Sound.

Source: Adapted from Valigura et al. 1996.

**TABLE IV-12**
**Studies of Atmospheric Loadings of Toxic Pollutants to NEP Coastal Waters**

| Coastal Water | Pollutants of Concern Evaluated[a] | Relative Contribution of Atmospheric Deposition for the Great Waters Pollutants of Concern | Reference |
|---|---|---|---|
| Massachusetts Bays (MA) | PAHs, PCBs, cadmium, lead, mercury | Direct atmospheric deposition estimated to contribute: PAHs, 9-46%; PCBs, 28-82%; cadmium, 17-31%; lead, 39-45%; mercury, 4-13%. | Menzie-Cura & Associates 1991 |
| Narragansett Bay (RI)[b] | PCBs, PAHs | Direct atmospheric deposition found to contribute 3% of PCBs and 12% of PAHs. | Latimer 1997 |
| New York-New Jersey Harbor Estuary and Bight (NY/NJ) | Cadmium, lead, mercury, PCBs, dioxins, PAHs, various pesticides | Atmospheric deposition identified as a significant contributor to total pollutant loading for lead (39-54%), but may have been over-estimated. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed. | NY-NJ Harbor NEP 1995 |
| Delaware Bay (DE/NJ/PA)[b] | Lead, mercury, PCBs, various pesticides, volatile organic compounds (VOCs) | Atmospheric deposition (both direct and indirect) found to be a significant source of mercury (80%) and PCBs (34%). For lead, atmospheric deposition contributed less than 5%. For other pollutants, either atmospheric deposition was insignificant or estimates were not developed. | Frithsen et al. 1995b |
| Tampa Bay (FL) | Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs | Direct and indirect atmospheric deposition identified as a significant contributor of cadmium (46%), lead (12%), and PCBs[c], but not a significant source of chlordane, DDT, dieldrin, or mercury (1% each). Estimates for PAHs were not developed. | Frithsen et al. 1995a |
| | Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs | On-going study - no results yet. | U.S. EPA 1995f |
| Galveston Bay (TX) | PAHs, PCBs, selected pesticides, lead, cadmium, mercury | On-going study - no results yet. | U.S. EPA 1995g |
| Santa Monica Bay (CA) | PAHs, metals, chlorinated organics | Atmospheric deposition was estimated to be a significant source of lead and PAHs. | SMBRP 1994 |

[a] For a discussion of other pollutants evaluated, study methods, and uncertainties, see referenced study.
[b] These NEPs also are NERRS designated sites.
[c] Estimates of PCB loadings could be made for atmospheric deposition only; therefore, a relative comparison to other sources could not be made.

A recent study of nutrient (i.e., nitrogen and phosphorus) and suspended solids loadings conducted for the Tampa Bay NEP suggests that direct atmospheric deposition of nitrogen to the tidal waters of Tampa Bay is the second largest source of nitrogen entering the Bay, contributing up to 28 percent of the total nitrogen load (Zarbock et al. 1994). The largest source of nitrogen, according to that study, is urban storm water runoff. A portion of the nitrogen entering the Bay from urban storm runoff represents atmospherically deposited nitrogen to impervious surfaces such as paved roads and sidewalks. These studies prompted the Tampa Bay NEP to revise its CCMP to consider atmospheric deposition issues.

Another study, conducted by the Tampa Bay NEP (Frithsen et al. 1995a), investigated the contribution of atmospheric deposition relative to point sources, urban runoff, and other nonpoint sources for specific chemical contaminants of concern. The contaminants, which were selected based on their potential for toxic effects and their concentrations observed in sediment samples, included: six metals (cadmium, chromium, copper, lead, mercury, and zinc); four organochlorine pesticides (chlordane, DDT, dieldrin, and endrin); and two classes of organic chemicals (polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs)). In addition, loadings were estimated for arsenic and iron because of the potential of these chemicals have for environmental harm or interaction with contaminants of concern through chemical processes. Because the study used numerous information sources, representing a wide range of spatial and temporal conditions, a great deal of uncertainty exists regarding the absolute estimates of atmospherically deposited chemical contaminants to Tampa Bay. Study results are intended to establish the relative magnitudes of different classes of sources in order to set priorities for more detailed research and monitoring activities. Some general conclusions, however, are apparent:

- Contaminant inputs to Tampa Bay from runoff and transfer of atmospherically deposited contaminants to the watershed (i.e., indirect loading) are approximately two-thirds the contribution of contaminant inputs from direct deposition to tidal waters (i.e., direct loading);

- Atmospheric deposition contributes a sizeable percentage to total annual load for the following metals: cadmium (46%), copper (18%), chromium (13%), lead (12%), and iron (11%);

- Atmospheric deposition of mercury is around 4 kg/yr and atmospheric deposition of pesticides is estimated as 10 kg/yr, each representing about one percent of the total load; and

- Atmospheric deposition is the only pathway that contributed a measurable amount of PCBs, estimated as a minimum total load of 11 kg/yr. No estimate could be developed for PAHs using available datasets.

Ongoing monitoring, described below, conducted by the Tampa Bay NEP, local governments, and other collaborators will better define the spatial distribution of atmospherically deposited chemical contaminants and nitrogen throughout the 10 major basins of Tampa Bay and its watershed as well as the relative contributions of local, regional, and global emission sources.

An ongoing cooperative study administered by the Tampa Bay NEP will substantially expand air transport and deposition monitoring and modeling projects in an effort to develop nationally recognized quantitative assessments for important air deposition parameters. Now in its second year of operation, the Tampa Bay Atmospheric Deposition Study (TBADS) involves EPA's Great Waters program; the local governments in Hillsborough, Pinellas, and Manatee Counties; the Florida Department of Transportation; the Southwest Florida Water Management District; Florida State University; a private consultant; and other organizations. TBADS will attempt to determine: (1) what fraction of the nitrogen and toxic pollutants emitted annually by specific sources within the Tampa Bay watershed enter the Bay waters (i.e., are deposited either onto the water surface directly or onto the watershed and subsequently enter the Bay waters through runoff); and (2) what are the relative contributions of local sources (i.e, inside the watershed) versus remote sources (i.e, outside the watershed) to atmospherically deposited nitrogen and toxic pollutants in the watershed. Projects that have been initiated to address elements of this air deposition program include:

- Intensive daily wet and dry deposition monitoring at Gandy Site, located on Tampa Bay's Interpeninsula, for nitrogen and toxic pollutants for an additional 12 months, yielding two years of continuous data collected and analyzed according to AIRMoN protocol;

- Application (with the Florida State University Center for Tropical Meteorology) of a regional air mass movement model developed by Pennsylvania State University (Penn State/NCAR Mesoscale Model Version 5) to investigate air transport at a coarse grid for the southeastern United States while maintaining a much higher resolution grid over the Tampa Bay area to estimate retention times for air masses within the watershed under different meteorological conditions;

- Integration of NOAA's Physical Oceanographic Real-Time System (PORTS) Meteorological data collected at several over-water stations (with NOAA and the University of Florida); and

- Intensive stormwater sampling to measure stormwater runoff, nitrogen, and toxic contaminant concentrations and loads at up to four gaged subbasins. Data will be used to estimate the relative contribution of atmospheric loading to stormwater for different land use types (residential, urban, industrial, and/or urban parks); it is expected that this transfer coefficient information will be useful not only to Tampa Bay and Florida, but also to watersheds nationwide.

An important element of the Tampa Bay atmospheric deposition program is the participation and coordination of local and federal government programs and state agencies with the Great Waters program and the Tampa Bay NEP.

**Galveston Bay**. The Great Waters program conducted a screening atmospheric deposition monitoring program in Galveston Bay, Texas, which was chosen as the site to establish monitoring for the Texas Regional Integrated Atmospheric Deposition Study (TRIADS) as a representative of a Gulf of Mexico estuary. Monitoring at the TRIADS site began in February 1995. To facilitate comparability, the sampling and analytical design of TRIADS is similar to that of existing monitoring sites in the Great Lakes and Chesapeake Bay. The goals of this study are

to evaluate the contribution of atmospheric deposition of selected contaminants to the Bay and to evaluate long-range transboundary transport of contaminants. Pollutants measured include cadmium, lead, mercury, nitrogen, PAHs, PCBs, and selected pesticides. Results from TRIADS complement and add to data from other investigations in Galveston Bay, including studies by the Galveston Bay NEP, EPA's Environmental Monitoring and Assessment (EMAP) program, NOAA National Status and Trends (NS&T) program, and special urban-pollutant studies in Houston, Texas. Data from these programs and the TRIADS data will be used to estimate the cumulative, direct and indirect impacts of atmospheric deposition to pollution of Galveston Bay.

Although early calculations suggested that atmospheric deposition could be a significant contributor to nitrogen loads delivered annually to Galveston Bay, the relevance of this finding to the health of the ecosystem for Galveston Bay is not as obvious as for either Chesapeake Bay or Tampa Bay. While all three estuaries have experienced declines in submerged aquatic vegetation (SAV), studies on SAV in Galveston Bay are limited in contrast to documented cases of large-scale changes in other major estuaries (Pulich et al. 1991). While atmospheric nitrogen loading may contribute to the incidence of hypoxia, other factors appear to be causing this phenomenon, which is quite localized in Galveston Bay compared to its manifestation in the other two estuaries.

In contrast to the perceived limited biological effects from atmospheric deposition of nitrogen, previous research has suggested that atmospheric deposition of toxic contaminants may be affecting fish and shellfish in Galveston Bay, and thus contributing to human health risk. A pilot study performed for the Galveston Bay NEP documented the presence of dioxins, furans, lead, mercury, PAHs, PCBs, and pesticides in certain species of finfish and shellfish, but could not determine the sources of these contaminants (Brooks et al. 1992). Monitoring data from the TRIADS site detected the presence of all these chemical contaminants in air samples, suggesting that atmospheric deposition may be a significant source (Battelle 1995). Continued monitoring will enable scientists and managers to more fully evaluate this problem and determine the relative effect of atmospheric deposition versus point and nonpoint source inputs into Galveston Bay.

**Casco Bay**. The primary pollutants of concern for atmospheric deposition to Casco Bay, Maine, include PAHs, PCBs, nitrogen, phosphorus, sulfates, pesticides, and mercury and other trace metals. Recent sediment studies have found elevated concentrations of some pollutants (i.e., cadmium, lead, mercury, PAHs, PCBs, silver, and zinc) near population centers and waste discharges, but also observed elevated levels in rural eastern Casco Bay away from these known sources (Wade et al. 1995). A circulation model study of the Bay did not clearly indicate any possible sources for these pollutants, suggesting atmospheric deposition as a significant source (Pearce et al. 1994). While elevated levels of lead found in Casco Bay sediments were relatively near potential sources, elevated levels of cadmium were found far from any known local source. A deposition study would provide empirical verification of processes believed to be occurring at Casco Bay.

**Delaware Bay**. In Delaware Bay, studies have shown that direct and indirect atmospheric deposition provide 15 percent of the annual nitrogen input, increasing to 25 percent in late spring and early summer (Scudlark and Church 1993). The relative nitrogen loading is slightly lower than observed in nearby Chesapeake Bay (27 percent), and much lower than in the Delaware Inland Bays (Rehoboth and Indian River Bays) where direct atmospheric deposition alone contributes 27 percent of the total nitrogen load (Cerco et al. 1994). The contribution to the Delaware Bay is lower because of higher point source nitrogen loading to Delaware Bay and the influence of a highly urbanized watershed.

As part of a Delaware Estuary Program study to estimate contaminant inputs, atmospheric deposition was found to be a significant source of mercury (80 percent) and PCBs (34 percent) (Frithsen et al. 1995b). As is the case in other regions, more research is warranted on atmospheric inputs of mercury and the resulting effects on estuarine and human health. To evaluate the effect atmospheric deposition of mercury has on the Delaware, Rehoboth, and Indian River estuaries, a precipitation monitoring station was established at Lewes, Delaware, in 1995 in conjunction with EPA's National Atmospheric Deposition Program Mercury Deposition Network.

**Gulf of Mexico**. It is probable that significant amounts of nitrogen are deposited into the Mississippi River Basin via atmospheric deposition, but there has been little investigation conducted regarding atmospheric nitrogen as a source of nitrogen for the Mississippi River drainage basin. Some basic estimates using National Atmospheric Deposition Program (NADP) data were provided at a Hypoxia Conference held by the Gulf of Mexico Program in August 1996. These estimates showed significant variability in quantity of atmospherically deposited nitrogen, with a range of 0.55 million to 3.08 million tons. This variability is due to differing assumptions of what atmospheric nitrogen input sources should be included, what forms of deposition should be measured (e.g., dry deposition), and what nitrogen compounds should be analyzed (e.g., ammonium). At the high end, atmospheric nitrogen would be on par with animal manure, ranking as the second highest source of nitrogen input to the watershed. At the lower end, estimated atmospheric nitrogen inputs would rank as the fifth highest source of nitrogen input for the watershed. This wide variability in estimated quantity points to the need for further and more refined estimates of atmospheric nitrogen inputs to the Gulf.

**Hypoxia Conference**

A conference was convened in 1996 in response to the hypoxia problem in the Gulf of Mexico. Topics addressed included:

- Characterization of the hypoxic zone;
- Economic impacts and trends in fisheries attributable to the hypoxia;
- Causes of the hypoxic zone;
- Sources and delivery of nutrients in the watershed, including atmospheric deposition;
- Current efforts to control nutrient loads; and
- Information and policy required for action.

**Long Island Sound**. A chronic problem in Long Island Sound is the low oxygen levels (hypoxia) that are observed during the summer. An early study noted that excess nitrogen loading was a major cause of hypoxia and estimated that atmospheric loading directly to the water surface contributed 8 percent of the total nitrogen delivered to the Sound (LIS Study 1990). A later study produced essentially the same estimate for the total contribution of direct atmospheric deposition, but divided it into two components: an amount from "natural" sources (i.e., background approximating "amount believed to have been delivered to Long Island Sound in pre-Colonial days") and an amount from "human-induced" sources (LIS Study 1994). Using measurements of wet and dry deposition from two sites along the Connecticut shore, Miller et al. (1993) estimated direct atmospheric loadings to Long Island Sound. The Long Island Sound Study used these data and literature values to develop the estimates shown in Table IV-12, and concluded that (direct and indirect) atmospheric deposition may be responsible for 17 to 24 percent of total nitrogen entering the Sound.

In the most recent study, all sources of nitrogen, including atmospheric deposition, are divided into "natural" and "human-caused" components (Stacey 1996). This study concluded that

atmospheric deposition from human activities in the New York and Connecticut portions of the Long Island Sound watershed accounts for 13.6 percent of the total enriched or "human-caused" load. Further work is necessary to model relationships among air quality, direct and indirect atmospheric deposition, and runoff concentrations to receiving waters of the Sound.

**Massachusetts Bays**. In one Massachusetts Bays NEP study, direct atmospheric deposition was estimated to contribute 5 to 16 percent of total nitrogen load to Massachusetts Bays (Menzie-Cura & Associates 1991). In another Massachusetts Bays NEP study, direct atmospheric deposition of nitrogen was estimated to account for 6 to 8 percent of total nitrogen loadings to the Bays (Zemba 1995). Different methodologies were used to estimate nitrogen loadings in these two studies. The estimate by Zemba (1995) used literature values and ten years (1981-1991) of wet deposition data from the NADP. Other studies cited by the Massachusetts Bays NEP suggest that the contribution of atmospheric deposition may be higher, about 16 to 20 percent of total nitrogen load, excluding exchange with the Gulf of Maine (Massachusetts Bays NEP 1996).

Atmospheric deposition is also a significant contributor of organic pollutants and trace metals to Massachusetts Bays. Menzie-Cura (1991) estimated that direct atmospheric deposition was a significant source of PAHs (9-46 percent), PCBs (28-82 percent), cadmium (17-31 percent), and lead (39-45 percent). A subsequent Massachusetts Bays NEP study generally corroborated the Menzie-Cura (1991) metal deposition results, although lead deposition rates were slightly lower (Golomb et al. 1995). The lead deposition estimates may be lower in Golomb (1995) because the data used in the Menzie-Cura study were obtained prior to the phase out of leaded gasoline. Golomb (1995) also indicated that PAH deposition may have been underestimated and that PCB concentrations were below detection limits and, therefore, atmospheric deposition rates for PCBs were not calculated. Because PCB concentrations were below the detection limit, more precise field measurements of wet and dry deposition of PCBs are necessary to verify the initial estimates and to determine the relative impact of atmospheric deposition of PCBs to Massachusetts Bays.

**Peconic Bay**. Nitrogen from atmospheric deposition to the Peconic River and Flanders Bay, New York, is estimated to be about 73 kg/day, or about five percent of the total nitrogen loading to that area (Suffolk County 1992). The impact of atmospheric deposition on eutrophication in Peconic River and Flanders Bay is considered to be relatively small in relation to other point and nonpoint sources. Atmospheric deposition is believed to be much more significant in terms of relative eutrophication impacts to Peconic Estuary surface waters east of Flanders Bay. Detailed loading estimates for these eastern areas, as well as for specific subwatersheds, are being developed for the Peconic NEP waters, and the relative eutrophication impacts of sources are being assessed through computer modeling.

## *Future Research Needs in NEP and Other Coastal Waters*

Research on atmospheric deposition to coastal waters has been limited to a few areas, and most studies have focused on identifying and determining the concentration of pollutants of concern in water and sediment, and measuring concentrations of pollutants in precipitation. Due to limited funding, many preliminary NEP studies are restricted to using historical data to estimate atmospheric deposition. Some NEP studies have used a rough mass balance approach

to determine the relative loading of each pollutant to the estuary, but more precise quantitative mass balances are needed, which require accurate and comprehensive atmospheric data.

Establishing the total contribution of pollutants and their sources is an important part of developing and implementing CCMPs for NEPs, and the lack of knowledge about the concentrations, deposition, and potential sources of airborne pollutants makes sound policy formation for the estuaries difficult. The question of the magnitude of pollutant deposition from the air has become more important as other sources of pollution to rivers, lakes, streams, and coastal waters have been identified and significantly reduced.

Research questions for the NEP estuaries and other coastal waters include:

- What are the concentrations and loadings of pollutants that are being supplied by atmospheric deposition?
- What are the relative contributions of these inputs to the total load of pollutants entering the estuary?
- What are the emission sources that affect the estuaries and where are they geographically located?
- Does atmospheric deposition (direct and indirect) of contaminants cause or contribute to biological harm in benthic (bottom-dwelling) or pelagic (suspended, planktonic, or water column) communities, or affect human health?
- What economically and technically feasible methods are available to effectively reduce airborne pollutants and their effects on estuaries?

The NEP estuaries provide an excellent opportunity to evaluate the effects and contribution of atmospheric deposition of contaminants to a varied set of ecological, environmental, and anthropogenic conditions. The NEP also provides a "grassroots" forum for addressing and correcting regional and national air quality issues as they pertain to our coastal waters. Recommendations for further atmospheric deposition research in coastal waters to help answer the above questions include:

- Utilize existing databases and ongoing work of established research programs and coordinate research initiatives with these programs;
- Protect and enhance existing monitoring programs;
- Establish long-term water and air quality monitoring programs that incorporate sampling for atmospheric deposition of contaminants for a subset of NEP estuaries representing various geographical regions and environmental conditions;
- Use sampling data from monitoring programs to track trends and spatial variability to develop more accurate loading estimates;

- Coordinate efforts between NEP estuaries and other Great Waters program studies to identify local, regional, and national sources of airborne pollutants;

- Pursue detailed atmospheric chemistry and deposition models for estimating atmospheric deposition to NEP estuaries;

- Develop a multi-party effort to identify and demonstrate appropriate pollution prevention techniques;

- Apply existing atmospheric circulation models to fill in data gaps between measured and estimated atmospheric deposition and to aid in tracing the pollutants in the estuaries back to their probable sources; and

- Support process-related research to establish cause and effect relationships between atmospheric deposition of contaminants and alterations of water quality, fisheries, recreational and other economic and ecological resources of receiving estuarine and coastal waters.

This research is needed not only to assist decision-makers for specific coastal waters, but to form a comprehensive picture of atmospheric deposition across the United States. In addition, coordinated use of other mechanisms, such as voluntary pollution prevention, can help control the negative impacts of atmospheric deposition to water quality in NEP estuaries, especially at the local and regional level.